Introduction

Positron Annihilation Lifetime Spectroscopy (PALS) has tremendous potential as a powerful tool for quantifying the types and densities of defects in solids. Following the injection of positrons into a solid material, the positrons pair-annihilate at a rate depending on the density of electrons near the injection site. If there are lattice vacancy or dislocation defects (voids) near the injection site, the positrons are attracted to these areas, which have lower electron densities and thus give rise to longer positron lifetimes. For conductor-type materials (metals) with many free electrons, the positron lifetimes can be quite short in defect-free regions (e.g. ~100 ps) and considerably longer in defect regions (e.g. ~200 ps). In the analysis of a lifetime decay spectrum acquired by PALS, the relative intensities of the various lifetime contributions that make up the spectrum can be extracted giving access to the density of defects near the injection site. Furthermore, if the timing resolution of the PALS system is precise enough, and the event statistics in the lifetime decay distributions are large enough, then information about the density of various types of defects could be extracted.

During the past quarter, continued development, characterization, and optimization of the ISU PALS apparatus has been performed by graduate student Brian Wieland with assistance from undergraduate student Kevin Tsai and ISU physics department assistant professor Dustin McNulty. The refinement of the apparatus is in preparation for two programs: The first involves validating the experimental apparatus' ability to accurately reproduce measurements of well-known positron lifetimes in high-purity samples using a radioactive ^{22}Na source. The second is a feasibility study and test of the Accelerator-based Gamma-induced Positron Annihilation Spectroscopy (AG-PAS) technique to perform the same lifetime measurements using either a Van de Graaff or Pelletron accelerator. Note that the main goal of this research is to exploit the technique of AG-PAS for nondestructive assessment of deep-defect densities in high-Z materials.

Source-based PALS measurements

In order to benchmark the PALS apparatus' ability to perform precision positron lifetime measurements, researchers have performed source-based measurements of positron lifetimes in high purity samples of Pb, Cu, Ni, and Al. While these measurements only acquired the minimum ~1M events per spectrum needed to separate the lifetimes in the sample from those in the source and backgrounds, the agreement of the data with well-known literature values are impressive. These preliminary results are shown in figures 1 – 5.

The spectra were collected under identical circumstances: the same source, geometry, system parameters, etc., and only varied in the material sample foils sandwiching the source. The detectors used were brand new Hamamatsu R3377 PMTs coupled to Saint-Gobain BC-418 fast plastic scintillators. The positrons were produced from a 5 year old, 50 μC ^{22}Na source encased in a very thin layer of Kapton tape. Five data sets, including source only with no sample, were simultaneously analyzed using the freely available LT10 fitting program [1]. Due to the identical data-taking conditions (only the sample varied), the simultaneous analysis allowed for powerful constraints on the characterization of the background lifetime components of Kapton and air (referred to as bkgd1), the source lifetime correction, and the timing response resolution of the system—all of which were common to each data-set. These common characteristics, as well as the individual sample parameters were freely varied to minimize the χ^2 for each data set as well as for all data-sets combined. It was found that the single Gaussian timing response function of the system had a Full Width at Half Maximum (FWHM) of ~255 ps, the source correction lifetime was 338 ps, the lifetime in Kapton was 370 ps, and the "bkgd1" lifetime contribution was 1.26 ns. Note that these results are preliminary and continued data-taking and analysis is still ongoing. Table 1 displays the fit parameters along with their relative intensities, and χ^2 for each data-set.

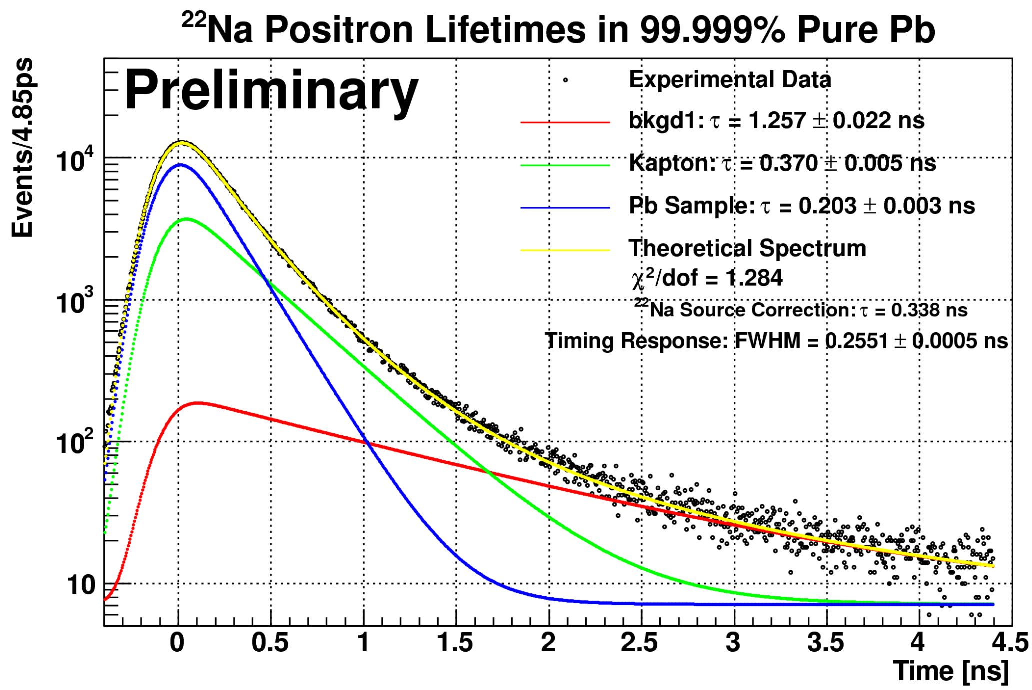

Figure 1: Positron lifetimes in Pb sample measured to be 203 ± 3 ps is in excellent agreement with the the literature value of 204 ps [2]. Note the poor statistics at larger times is a result of only acquiring ~1M events. Ideally, ~5M events minimum should be acquired for these spectra.

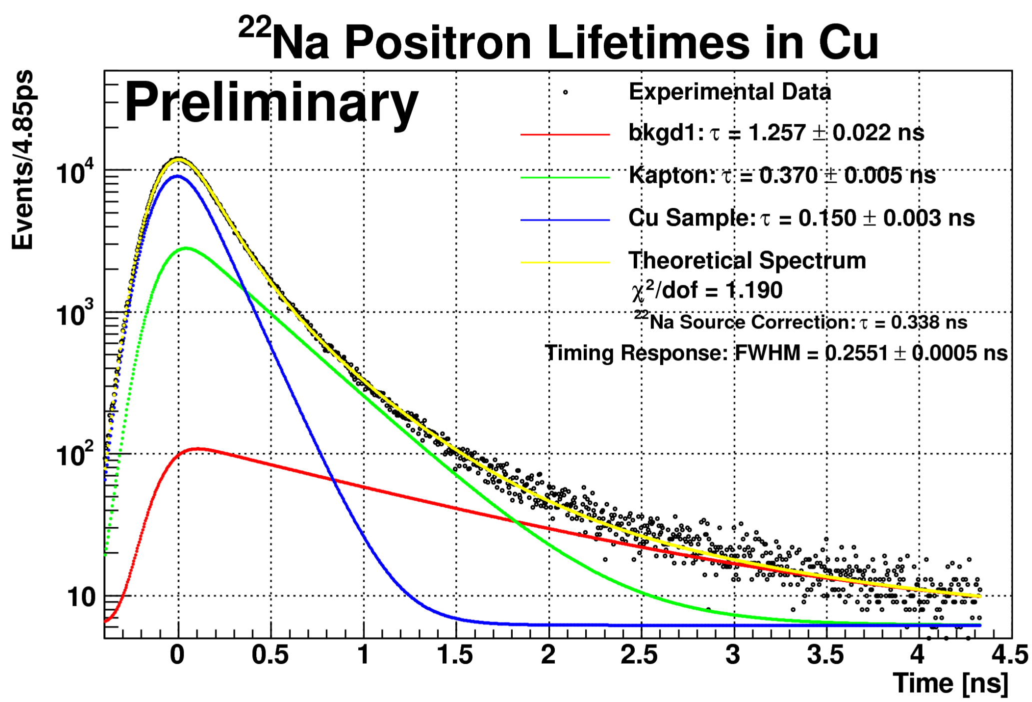

Figure 2: Positron lifetimes in Cu sample measured to be 150 ± 3 ps. The Cu foil sample used was the only sample which had unknown purity.

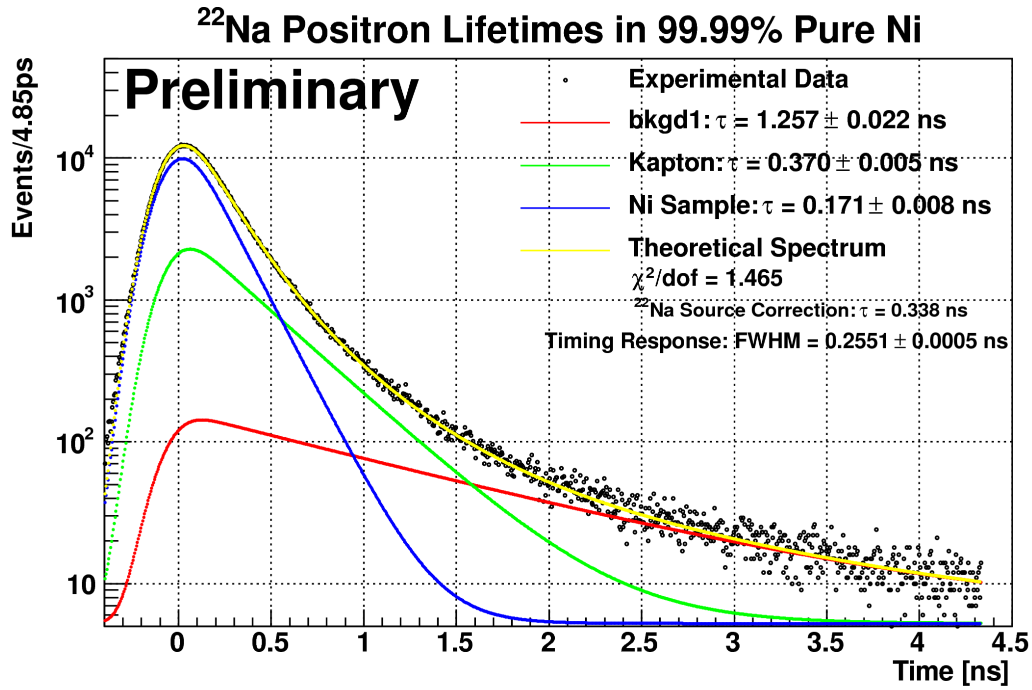

Figure 3: Positron lifetimes in Ni sample measured to be 171 ± 8 ps. Note that the bkgd1 lifetime parameter common to all sample data-sets is thought to be from positron decays in the thin layer of air between the Kapton encasing the source and the PMT scintillator.

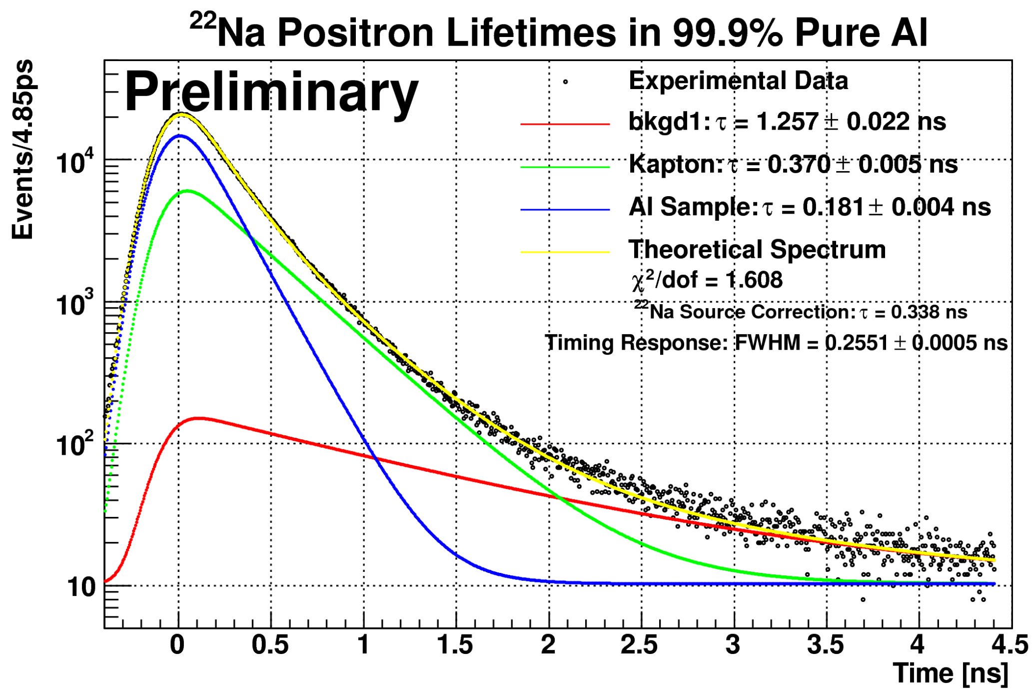

Figure 4: Positron lifetimes in Al sample measured to be 181 ± 4 ps. It can be seen from Table 1 that the intensities of the Kapton lifetime component are very similar for all data-sets, as expected.

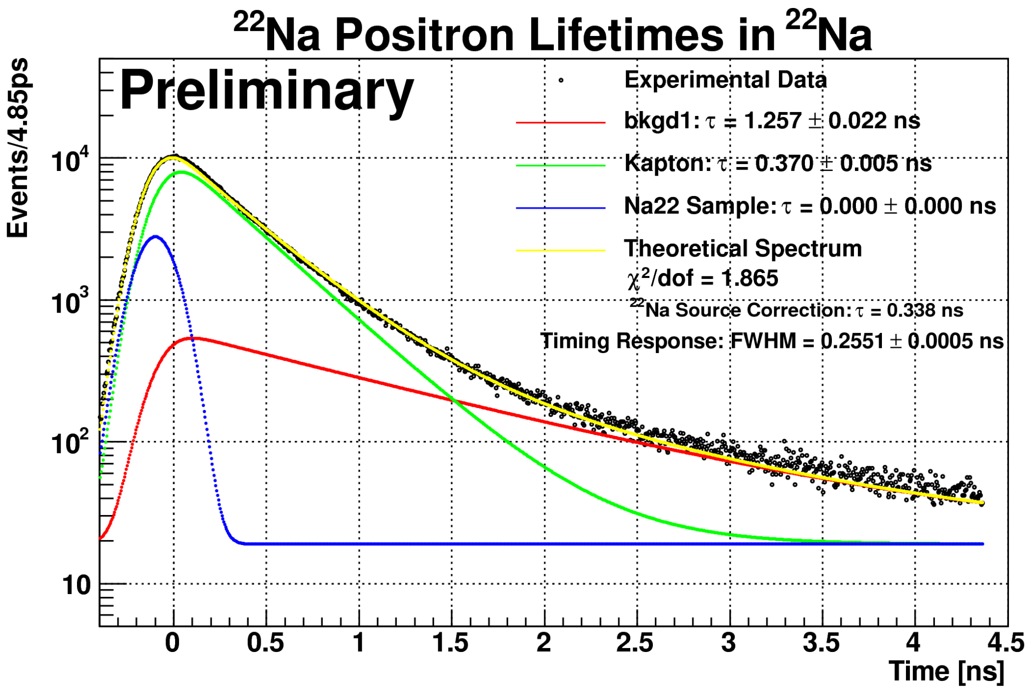

Figure 5: Positron lifetime in the source itself. The sample lifetime (blue) curve displays the actual timing response function (single Gaussian) which is convoluted with the exponential decay distributions for all data-sets. More statistics and analysis are needed for this distribution as it gave the worst χ2 and is perhaps the most important spectra to fully characterize since it is common to all data-sets.

Table 1: Fit parameters from source based PALS measurements

Sample	Intensity1 (%)	τ1:bkgd (ns)	Intensity2 (%)	τ2: Kapton (ps)	Intensity3 (%)	τ3: Sample (ps)		Fit χ^2/d.o.f
Pb	4.24±0.00	1.257±0.022	35.0±2.99	369.9±4.74	60.7±4.20	202.7±2.58		1.2839
Cu	2.90±0.00	1.257±0.022	32.0±1.79	369.9±4.74	65.1±1.09	149.9±3.12		1.1896
Ni	3.71±0.00	1.257±0.022	24.7±4.87	369.9±4.74	71.5±6.21	170.9±7.91		1.4649
Al	2.13±0.00	1.257±0.022	36.7±1.97	369.9±4.74	61.1±3.16	180.9±3.56		1.6079
^{22}Na	12.3±0.00	1.257±0.022	76.0±0.85	369.9±4.74	11.7±0.82	0.0±0.00		1.8653

As mentioned, these results are preliminary, but are very promising. Future source-based measurements will acquire greater event statistics. This will enable additional sample lifetime parameters to be extracted. While the above analysis only allowed for a single sample lifetime parameter (τ3), future measurements will allow for multiple sample lifetime parameters to facilitate a quantitative assessment of bulk (defect-free) versus mono-vacancy surface densities. Table 2 gives some recent literature values of experimental measurements of positron lifetimes to compare with the ISU source-based PALS measurements. Literature values are given for both bulk and mono-vacancy sample measurements from two different references [2, 3]. All four ISU measured samples, of unknown defect densities, are in agreement with these reference values. It is perhaps spectacular that the Pb sample agrees very closely with the bulk reference value which is what one would expect since Pb is self annealing. The Cu, Ni, and Al samples produced single lifetime parameters in-between the bulk and mono-vacancy values which is precisely what one would expect from off-the-shelf, non-annealed samples of unknown defect densities.

Table 2: Experimental lifetime values from recent literature compared with ISU measurements.

Sample Material	Experimental Lifetime Values from Literature (ps)				ISU PALS measurements (ps)
	Bulk (no defect)		Mono-vacancies		Unknown Defects
	ref[2]	ref[3]	ref[2]	ref[3]	
Pb	204	194	294	No data	202.7 ± 2.58
Cu	120	100	180	173	149.9 ± 3.12
Ni	109	No data	180	No data	170.9 ± 7.91
Al	165	163	244	244	180.9 ± 3.56

<u>Accelerator-based PALS measurements (future plans)</u>

Following the source-based validation of the PALS apparatus, researchers will perform accelerator-based, gamma-induced positron lifetime measurements. Of course the main advantage of this technique lies in its ability to probe the volume of a sample as opposed to only the surface. The initial goal is to study the feasibility of performing these measurements, and in particular, map-out the optimal geometry between Aluminum proton-capture target, sample, and detectors, as well as explore various shielding/collimation configurations of the detectors and beam. The key parameters under investigation in these initial measurements will be the size and signal to noise ratio of the overall coincident rate of lifetime events as a function of geometry, shielding, beam current, and sample size.

Initial measurements will be done using the Van de Graaff accelerator which is capable of producing a ~10 μA, 1.5MeV proton beam. However, since the PALS measurements require a coincidence between two detectors, combined with the isotropic nature of both the “start” and “stop” events, the rate of lifetime events may be too low to acquire the necessary statistics in a reasonable amount of time. For this reason, the Pelletron accelerator which is capable of ~200 μA, ~8 MeV will need to be used. Additionally, the proton capture reaction on Aluminum ($^{27}Al(p,\gamma)^{28}Si$) used to produce the photons necessary for the measurement may simply have too low of a photon yield—the cross section for the capture reaction is fairly low for incident protons below 10 MeV. Researchers therefore may need to explore other proton capture reactions or other means of positron production to achieve the coincidence rates necessary to facilitate sample lifetime measurements in a reasonable amount of time. These measurements will go into full production in the coming weeks and continue throughout summer 2012.

References

[1] Kansy, Jerzy, and D. Giebel. "Study of defect structure with new software for numerical analysis of PAL spectra." *J. Phys.: Conf. Ser.* **265**, 1, 2011.

[2] J M Campillo Robles, E Ogando, and F Plazaola. "Positron lifetime calculation for the elements of the periodic table." *J. Phys.: Condens. Matter* **19** 176222, 2007.

[3] P.A. Sterne and J. H. Kaiser. "First-principles calculation of positron lifetimes in solids." *Phys. Rev. B:* **43** 13892, 1991.