Development and testing of a three detector setup for PALS measurements

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Abstract of the bachelor's thesis

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Abstract

While Positron Annihilation Lifetime Spectroscopy (PALS) is a very effective method for measuring the presence of vacancy-type defects in a material structure, its reliance on detecting photons to calculate the lifetime of a positron means that, in the case of a radioactive sample, the lifetime spectra get distorted by this additional radiation. A solution to this problem is to use three detectors instead of the traditional two: one for detecting the start gamma and two for the annihilation gammas [1]. This would eliminate the majority of the false counts that cause the distortion, because unlike annihilation which always produces two simultaneous and co-linear photons, radioactive decay is often isotropic.

In order to allow better handling while taking the necessary measures for dealing with radioactive materials, a new source was needed. This source was prepared by enclosing ^{22}Na salt between two layers of Kapton foil, with a sturdy plastic ring and a special sample holder to make it easier to handle. It was of course necessary to demonstrate that this new prototype source could be used for lifetime measurements by running traditional two detector PALS measurements with it and comparing the results to those of usual sources. The activity of the source and the necessary source corrections for the eventual analysis were also determined as a part of this process.

After the source had been validated, the next step was to optimize the detectors. The biggest challenge in this was optimizing the time resolution, for which the Constant Fraction Discriminator (CFD) threshold response spectrum would need to be obtained. However, the system being not equipped with a Multi-Channel Analyzer (MCA), an innovative and elegant solution was proposed to reconstruct the energy spectrum of a given radioactive source. With the CFD threshold response spectrum, an optimized time resolution function could finally be obtained using a 60Co source.

Finally, once the source had been prepared and validated and the detectors optimized, the entire setup was tested to demonstrate it could eliminate the distortion caused by radiation from the sample and determine the device performance. Ultimately, a significant improvement was achieved with the three detector setup compared to using just two detectors.

Keywords Positron Annihilation Lifetime Spectroscopy, Triple-coincidence, Radioactivity

Preface

I want to thank Professor Filip Tuomisto for taking me into the Positron group for one of the best summers of my life. I also want to thank Doctor René Bès for his excellent guidance, both during the time I was in the Positron group, and with helping me bring this thesis to its conclusion all these years later. I would also like to thank Senior University Lecturer Jani-Petri Martikainen for helping me finish this thesis. And finally, I would like to thank all my friends and family who supported me throughout all these years.

Helsinki, 21.01.2025

Toni S. Simola

Contents

1 Introduction

The energy needs of the modern world are rising every year. The vast majority of this energy is produced using fossil fuels, which not only will be mostly depleted over the coming decades, but more importantly, also pose a great threat to the environment. Nuclear power, while also not a renewable source of energy, will last much longer and has much less severe consequences for the environment when properly managed. Most importantly, the carbon-free nature of nuclear power means that replacing the use of fossil fuels with it would allow us to greatly limit global warming. As such, nuclear power presents a potential solution to the current climate and energy crisis.

A better understanding of nuclear fuel behavior can greatly help in the development of safer and more efficient methods of nuclear power. Irradiation can form vacancy-type defects (an atom missing from a lattice) in fuels that can eventually evolve into larger voids and dislocation loops. The structural components of nuclear reactors can also receive similar radiation damage.

Positron Annihilation Lifetime Spectroscopy (PALS) offers a method of studying these vacancy-type defects in solids. PALS works by trapping positrons inside matter and measuring the lifetime of the positron, as that correlates with the amount and nature of vacancy-type defects in the studied sample. However, traditional PALS methods cannot be used for studying radioactive materials, such as nuclear fuels or irradiated steels, because $\beta+$ and gamma radiation interfere with the measurement of positron lifetimes by producing false coincidence events, greatly decreasing the accuracy of PALS measurements. Currently, PALS has mainly been used to measure low-activity materials (below 100 kBq).

A new Triple-Coincidence Positron Lifetime Spectrometer was proposed by Janne Heikinheimo, René Bès, and Filip Tuomisto in their paper *Development of a Triple-Coincidence Positron Lifetime Spectrometer for Nuclear Materials Research* as a possible new PALS setup that would enable the studying of radioactive materials [1]. In this manuscript, I will describe the development, building and testing process of this new Triple-Coincidence PALS setup.

The development process consisted primarily of three major steps. First, the setup was built, and the detectors were calibrated and optimized for use in actual measurements. Then a new source was prepared and validated for use in the setup. Finally, the complete setup was tested in a simulated radioactive environment, where radiation sources were placed in close proximity of the measured sample to demonstrate that the setup could be used to study radioactive materials.

2 Positron Annihilation Lifetime Spectroscopy

2.1 The basic principles of PALS

Positron Annihilation Lifetime Spectroscopy (PALS) is an experimental technique used primarily for finding vacancy-type defects in the lattice of solid materials. A positron source is placed between two samples of the material we wish to measure. This positron source is made up of a radioactive isotope, usually ^{22}Na, that decays through β^+-decay, where one proton in the nucleus transforms into a neutron and emits a neutrino and a positron. As ^{22}Na decays, it turns into ^{22}Ne, which is left into an excited state, which collapses essentially immediately, emitting a photon with the energy of 1.27 MeV. This photon is then detected by a nearby detector, and since the emission and subsequent detection of the photon happen so quickly that it's almost simultaneous with the emission of the positron from the β^+-decay, this can be used to mark the start of the positron's lifetime.

This positron then travels to the samples surrounding the source, where it eventually annihilates with an electron, converting both of their masses into two photons with an energy of 511 keV each. One of these photons is then detected by another detector which tells us the exact time of the annihilation. Knowing the emission and annihilation times, we can now calculate the lifetime of the positron. A schematic overview of a standard 2-detector PALS setup can be seen in figure 1. [3]

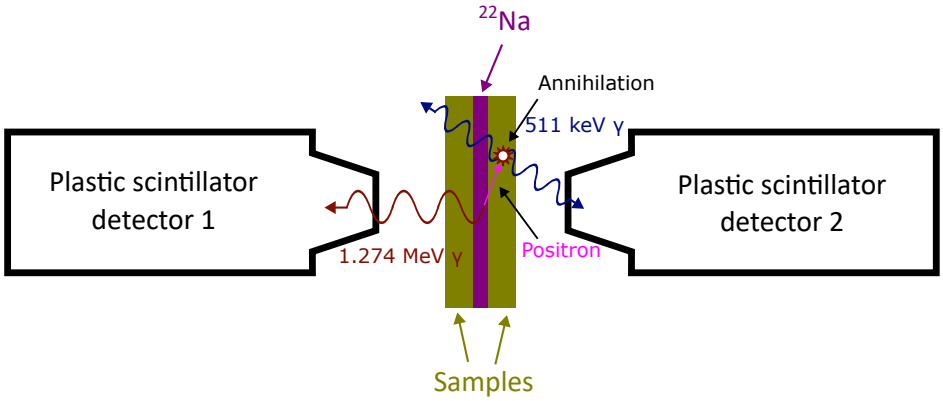

Figure 1: A basic schema of a standard 2-detector PALS setup. A positron source is placed between two samples to form a "sandwich". Two plastic scintillator detectors are placed in opposing sides of the sandwich with detector 1 calibrated to detect the *start* gamma that coincides with the emission of the positron, and detector 2 to detect the *stop* gamma from the eventual annihilation.

PALS is a statistical technique, thousands of positron lifetimes are counted over a period of time ranging from a couple of minutes to days and then the results are compiled to a "lifetime graph" to show the total distribution of positron lifetimes. These lifetimes are extremely short, between 100–400 picoseconds in solids [1]. How

long a PALS experiment lasts largely depends on the count rate of the setup, i.e. how many lifetime events are recorded per second, and the count rate is affected by the different factors, such as the activity of the positron source, the solid angle between the detectors and the source, as well as the time resolution and detection efficiency of the detectors.

Positron lifetimes can reveal us a lot about the amount and nature of vacancy-type defects within the sample. As charged particles, they are repelled by the positive nuclei in matter. This causes them to be drawn to vacancies where they achieve a lower energy state due to being farther away from the nuclei, which then results in a smaller likelihood of annihilating with electrons, extending the lifetime of these positrons. Therefore, there is a correlation between the lifetime graph and the nature and amount of vacancy-type defects in a sample.

There are two main types of defects that can be formed within a lattice: extended defects and lattice point defects [2]. Extended defects are larger scale defects consisting of multiple atoms, while lattice point defects are small and only occur in a single lattice point. While there are a total of 5 types of lattice point defects (vacancy-type defect, interstitial defect, Frenkel pair, antisite defect and substitutional atom), PALS is mainly suited for studying vacancy-type defects due to how they interact with positrons as explained above. In a vacancy-type defect, an atom or an atom cluster is simply missing from a lattice, leaving a "hole" in that lattice.

2.2 The standard two detector PALS setup

The standard PALS setup consists of two detectors facing each other with the source and sample between them. The detectors are placed as close to the sandwich as possible to maximize the solid angle to achieve a better count rate [3].

The source is carefully placed between two samples of the material being studied, and this is typically called a *sandwich*. The source itself is β^+-active material, typically ^{22}Na salt, folded inside a metal foil (usually aluminum or nickel). Many of the positrons emitted by the ^{22}Na salt will of course annihilate in this foil, which is why it is made to be as thin as possible to limit this, while still keeping the salt contained inside of it to avoid contamination of the equipment and the samples. The basic principle behind this sandwich is to place the samples as close to the source as possible to maximize the amount of positrons that annihilate in the sample.

One of the detectors is calibrated to detect the 1.27 MeV *start* gamma and the other one to detect the 511 keV annihilation *stop* gamma and are typically called *start detector* and *stop detector* respectively. The detectors are connected to a computer where the data is collected and combined into the lifetime graph.

3 The Three Detector PALS Setup

3.1 Theory and introduction

PALS has been a very successful and effective technique for finding vacancy-type defects in solids, but is greatly limited by requiring the studied sample to be low

activity (below 100 kBq) [1]. In particular, PALS would be a valuable tool for nuclear fuels and reactor structures. As noted by Janne Heikinheimo, René Bes and Filip Tuomisto in references [1] [2], in nuclear reactors, vacancies can form from high-speed collisions with other particles and are the primary radiation damage that eventually evolves into larger voids and dislocation loops. Vacancy-type defects have an important role in determining multiple properties, such as radiation tolerance, diffusion of atoms in the material, and mechanical strength and endurance, in both the nuclear material and the structural components of the nuclear reactor itself.

Typical PALS methods are limited to studying low-activity materials because the photons emitted by radioactive samples interfere with the measurement process. These photons are picked up by the detectors that are supposed to detect the *start* and *stop* gammas, and since random photons from the sample are indistinguishable from the photons emitted through positron emission and annihilation, they're falsely recorded as lifetime events. Not only does this increase the amount of background radiation present in the experiment, it will also distort the shape of the lifetime graph, making it impossible to analyze to determine the positron lifetime components from the sample [1].

In order to only count real lifetime events, there needs to be a way to distinguish them from the false events caused by the radiation from the sample itself. While this radiation is unlikely to produce photons with exactly 1.274 MeV or 511 keV energy, detectors cannot be tuned precisely enough to only detect very specific energy ranges without losing the necessary timing accuracy for lifetime measurements (the commonly used plastic scintillator detectors tend to have very poor energy resolution because of this). A better way to only count real lifetime events is to include a third detector to the setup, which is also set to detect the annihilation photons. Radioactive decay is random, but an annihilation will always result in two photons emitted in opposite directions. While special modes of decay can produce three or more, or just one photon, these cases are rare enough to be irrelevant in usual PALS measurements [4]. Hence, if two simultaneous photons of 511 keV are detected in coincidence with the 1.27 MeV photon, it can be concluded to be a real lifetime event. Of course, radioactive decay will still randomly produce false events even in triple coincidence, but those are exponentially less likely, making the resulting error negligible.

Similar 3 detector PALS setups have already been proposed and built in the past, particularly for measuring irradiated steel [7] [8]. The particular setup we built in this thesis was based on the study by Janne Heikinheimo in reference [1], with the main goal of researching spent nuclear fuel.

3.2 The experimental setup

The three detector setup is almost the same as the standard two detector setup, with the obvious addition of an extra detector. Two stop detectors are placed facing each other on either side of the sandwich and the start detector is placed perpendicular to them. The stop detectors need to be facing each other because in the event of an annihilation, photons are emitted in opposite directions. In the case of the "start

detector", simply the surface area needs to be maximized. A schematic overview of the three detector setup can be seen in figure 2.

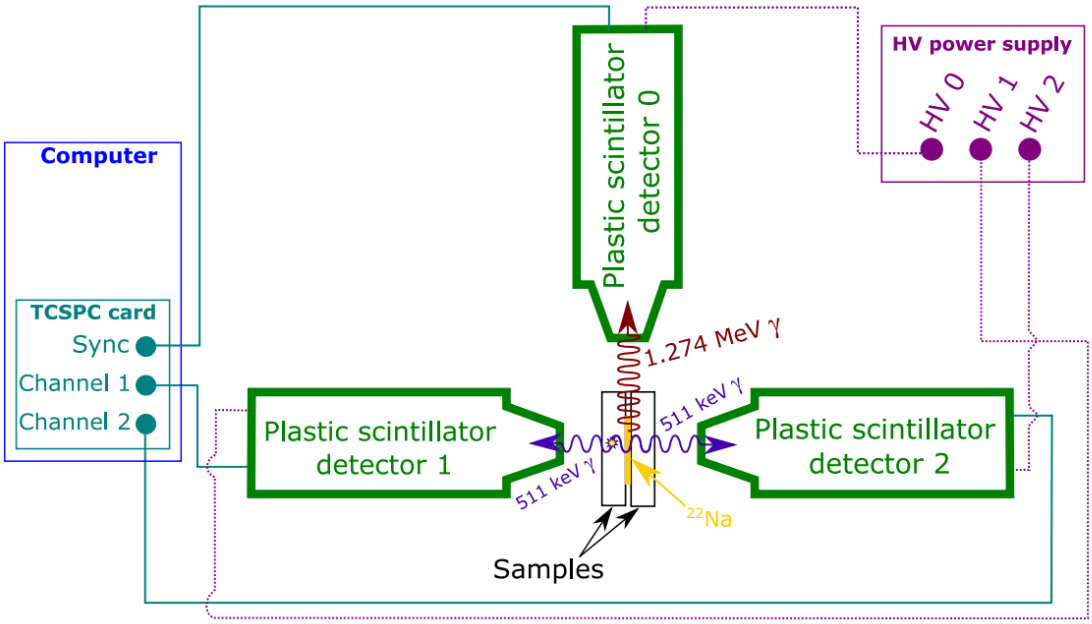

Figure 2: A schematic overview of the 3 detector setup, as well as the wiring with the HV power supply and the TCSPC card. Image source [5]

The three detectors used were plastic scintillator detectors because they allowed for better timing efficiency. The detectors were then connected to a computer with three 50 Ω coax cables. The computer (Windows 7 64-bit OS, Intel Xeon CPU dual core at 3.20 GHz and 8 GB of RAM) was equipped with a Time-Correlated Single Photon Counting (TCSPC) PCIe-card that read the input signals from the detectors. The detectors were powered by connecting to a basic power supply with three SHV cables.

Because the event of a triple coincidence is significantly less likely than a double coincidence, the count rate of a three detector setup is expected to be much lower. The count rate also suffers from the fact that it's not possible to fit three detectors as close to a sample as two detectors, which reduces the solid angle of the photon detection. These are partially mitigated by the use of a more powerful source (higher source activity leads to more positrons and consequently more annihilation events to detect), but it still presents a challenge that needs to be overcome through optimization of the resolution function. While a speed similar to standard 2 detector setups is likely not possible, the measurement still needs to be relatively fast, not only for convenience, but also because radioactive materials decay over time, which might alter the sample itself. The sample could end up being significantly different by the end of an extremely long measurement.

While this was the experimental setup used for our testing and development,

there are two notable differences between it and the final concept. The first was the orientation of the detectors, in this experimental setup they all lay on a table horizontally, but in the final setup, the two *stop detectors* would be mounted vertically above and below the sample and only the *start detector* would be horizontal. The detectors would also be mounted on rails to more finely tune their distance from the sandwich and ensure similar distance with both *stop detectors*. This vertical mounting would allow for more convenience in real measurements as the sample holder could simply be slid between the detectors. However, this did not matter for testing purposes, and we chose to keep the detectors horizontal where they could simply lay on a table instead of needing to build a structure to hold them vertically. Their positions were adjusted by hand and distances to the source measured rather crudely with a basic ruler.

The second difference was lack of the glovebox. When analyzing highly radioactive materials, the setup would need to be operated inside a glovebox for safety reasons. Again, this was mainly omitted for convenience, as none of the radioactive materials we used for testing were dangerous enough to necessitate safety measures beyond basic lab coats, gloves, pincers and goggles.

3.3 The recording of lifetime events

The detectors work by sending an electric pulse whenever they detect a photon, with this pulse height proportional to the energy of the photon. Since we need a very precise measurement of the time of detection, recording the timing of a lifetime event when this output pulse reaches a certain height, would cause problems as the stronger pulses from the detection of the 1.28 MeV *start* gamma would be detected faster than the 511 keV *stop* gamma.

Because of this, a Constant Fraction Discriminator (CFD) is used. A CFD records the timing of a pulse when it reaches a constant fraction of the pulse's maximum height. This eliminates the dependency between pulse height and the recording time.

Each detector simply logs all the times when they detect a photon, and these logs get compared against each other to determine positron lifetimes. In a standard two detector system, if a *start* and a *stop* gamma are detected within a time gate t_{gate}, it is recorded as a lifetime event. This time gate exists to cut out scenarios where a *stop* gamma simply missed the detector, but good practice is to set it to be at least 10 times longer than the expected positron lifetime to ensure all real lifetime events are recorded.

In a three-detector setup, there are two time gates: a long time gate $t_{gatelong}$ and a short time gate $t_{gateshort}$. The long time gate is the same as the regular time gate in a 2-detector setup. The short time gate requires that the two *stop* gammas are detected within that time gate or a lifetime is not counted. This short time gate is set to be long enough to account for the resolution of the detectors, but short enough to limit false counts as much as possible.

3.4 New sandwich for three detectors

A traditional positron source for PALS measurements has ^{22}Na salt enclosed inside a very thin, very fragile foil of aluminum. This presents a problem as one of the necessary safety measures is a glovebox, that makes both preparing and handling such small and fragile objects cumbersome, if not outright impossible. Although our experimental setup did not include a glovebox, we still needed to create and test a new type of positron source that was bulky and sturdy enough to be used in such an environment for future use.

The new prototype source, dubbed "Proto1", would have the ^{22}Na salt enclosed within two 8 μm thick kapton foils. These foils would be held together by two polyether ether ketone rings that would prevent the salt from leaking away from between the kapton foils, and to provide the necessary bulk and durability to the source. The studied samples would be placed inside the ring, where they would be in direct contact with the kapton foil (and consequently extremely close to the ^{22}Na salt itself) to allow for most of the positron annihilations to happen in the sample, like with a regular PALS source. A schema of the source can be seen in figure 3.

Another reason why a new source was needed was because of a major weakness of the 3-detector PALS setup is expected to be its low count rate. Having a more powerful, higher activity source would help to mitigate this weakness as the count rate is proportional to the activity of the source. In typical PALS setups, higher activity sources can be problematic as the source itself is the main cause of "background" radiation in a measurement. However, as the 3-detector setup is specifically designed to reduce the effects of background radiation, this is not a problem.

^{22}Na has a half life of 2.6 years and usually PALS sources can be expected to last roughly 6-10 years [3]. Proto1 has higher-than-average activity, but since this high activity is supposed to compensate for the expected low count rate of the three detector setup, it most likely needs to be replaced at shorter intervals than regular PALS sources.

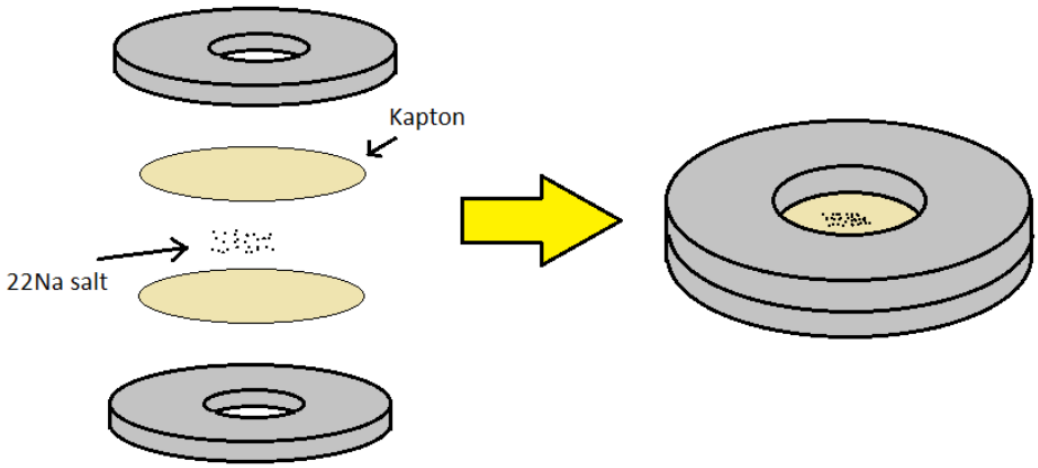

Figure 3: Schematic representation of the Proto1 source. of ^{22}Na salt is encased between two thin layers of kapton foil, with two PEET rings keeping it sealed.

To make the source, we mixed 1 ml of distilled water into a bottle of 3.7 MBq of ^{22}Na salt. The resulting mixture was then pipetted one droplet at a time on top of a small piece of kapton foil and the water was allowed to vaporize, leaving the ^{22}Na salt stuck on the foil. The droplets vary slightly in size but were estimated to be about 2 mm in diameter. This process was repeated a total of 18 times when making the Proto1 source. The salt was then sealed using a second sheet of kapton foil and the PEET rings.

The source also had a special sample holder to add more bulk to the sandwich while keeping the samples and the source together. The sample holder consisted of two rectangular PEET plates held together by magnets, with a small cavity between where Proto1 and the samples would be placed.

3.4.1 Determining the activity of Proto1

The next step was to measure the activity of Proto1. While there are multiple methods for measuring the activity of a positron source, we chose to do that by using a regular two detector PALS setup. You can run a basic PALS measurement of the source itself without any sample and determine the activity based on the results as the number of coincidence events counted by the measurement is directly proportional to the activity of the source.

One might assume that the geometry dependence of a PALS measurement might be a hindrance, as slight changes in the distance between the detectors and the source or their angle can also affect the count rate. However, knowing the ratio between true coincidences and random coincidences measured by the setup, this geometric

dependency can be eliminated. The number of true coincidences detected (N_{true}) depends on the activity of the source (A_{source}) and these geometric variables:

$$N_{true} = A_{source} \times \epsilon_0(1274)\frac{\Omega_0}{4\pi} \times \epsilon_1(511)\frac{\Omega_1}{2\pi}, \tag{1}$$

where $\epsilon_0(1274)$ and $\epsilon_1(511)$ are the efficiencies of detectors 0 and 1 at detecting the 1.247 MeV *start* and 511 keV *stop* gammas respectively. Ω_0 and Ω_1 are the solid angles of the detectors, the source of this geometric dependency that we wish to eliminate.

The number of false random coincidences (N_{random}) is also dependent on the geometry of the setup. The main source of random coincidences is from two decays in the ^{22}Na source happening in very close proximity to each other; within the same time gate (t_{gate}). This can result in the *start* gamma being detected from one decay and the *stop* gamma from another. As the detectors only have a lower threshold, it is also possible for the 1.247 MeV gamma from the second decay to be counted as a *stop* gamma by detector 1. False coincidences can also be caused by random background radiation, either between two gammas from the background, or in coincidence with one gamma from the source itself. However, since the activity of the source is significantly higher than the background, this can be considered negligible. Thus, we can write:

$$\begin{aligned} N_{random} \approx\ & A_{source}^2 \times \epsilon_0(1274)\frac{\Omega_0}{4\pi} \times \epsilon_1(511)\frac{\Omega_1}{2\pi} \times t_{gate} \\ & + A_{source}^2 \times \epsilon_0(1274)\frac{\Omega_0}{4\pi} \times \epsilon_1(1247)\frac{\Omega_1}{4\pi} \times t_{gate} \\ & = A_{source}^2 \times \epsilon_0(1274)\frac{\Omega_0}{4\pi} \times \left(\epsilon_1(511) + \frac{1}{2}\epsilon_1(1247)\right)\frac{\Omega_1}{2\pi} \times t_{gate}, \end{aligned} \tag{2}$$

where $\epsilon_1(1247)$ is detector 1 efficiency at detecting 1.247 MeV gamma. We can assume that $\epsilon_1(511) \approx \epsilon_1(1247)$ i.e. that both types of gammas are detected at the same efficiency. Therefore, we can take the ratio of N_{random} and N_{true} to eliminate the geometric variables to get:

$$\frac{N_{random}}{N_{true}} = \frac{3}{2}A_{source}t_{gate}. \tag{3}$$

To determine the source activity, a 15-hour PALS measurement was preformed. The time gate was set to 6 ns and the total count rate was 91.8 counts per second. The resulting lifetime spectrum can be seen in figure 4.

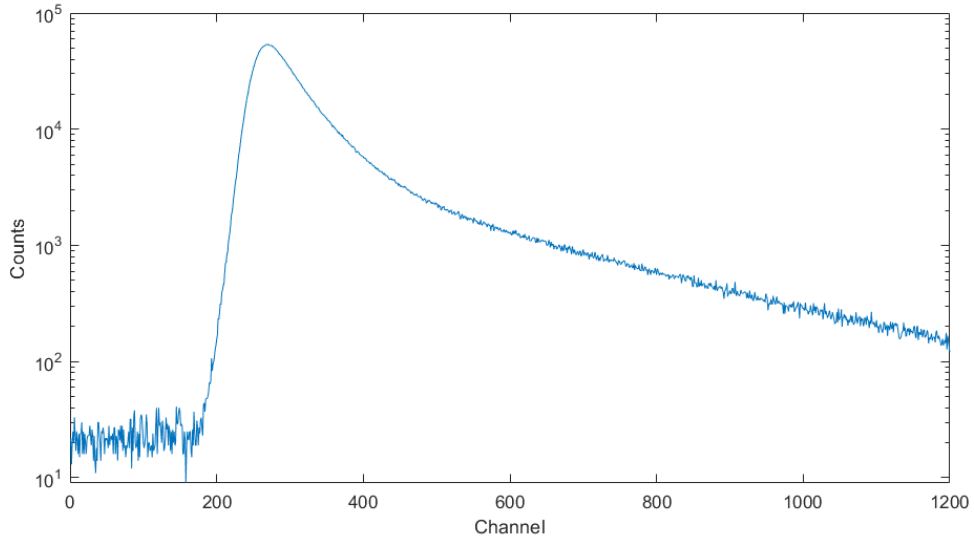

Figure 4: Measurement data from Proto1 activity measurement. Each channel is a time interval of 8 ps.

The peak is caused by real lifetime events N_{true} while the random false coincidences N_{random} make up the background noise in the graph. We can estimate N_{random} by taking the average number of counts per bin in the first 150 bins, i.e. before the peak where no real lifetime should be, and multiplying this average by the number of bins on the x-axis. N_{true} is of course the total number of counts N_{total} minus the random false counts N_{random}. The values of N_{true} and N_{random} were determined to be $4.93e+6$ and $27.1e+3$ respectively, and using formula 3 the source activity was determined to be 0.6(1) MBq.

3.4.2 Determination of the source correction parameters

The lifetime spectrum is made up of multiple lifetime components in addition to the background. A significant number of positrons emitted by the source annihilate in the source itself or form positronium states near the surfaces of the source or the sample, and all of these leave their characteristic lifetime components to the lifetime spectrum. These lifetime components left by the source are called source correction parameters and need to be accounted for in the data analysis process to get the true lifetime of the sample itself.

In order for Proto1 to be usable for PALS we then need to figure out its source correction parameters. There are a total of three lifetime components caused by the source that all have their own correction parameters. The first one of these is from annihilations in the 22Na salt itself, the second one from annihilations in the kapton foil and the third one from long-lasting positronium states. For each of these correction parameters, we need to know the average positron lifetime in that material, as well as the intensity of this lifetime component in the results, i.e. the percentage of how many positrons annihilate in that part of the source.

Determining the lifetime components is relatively simple. We can take a sample with a known positron lifetime component and measure this sample with Proto1. As we already know the resulting lifetime from the sample, we can subtract it from the measurement results and treat the source correction parameters as the unknown lifetime to be measured. For this process we used the PALS data analysis software PALSfit [6], which allowed us to set and tweak source correction parameters for any PALS measurement.

The lifetime component of the kapton foil is the easiest one to figure out. It has been measured that the positron lifetime in kapton foils is 385 ps [10], so all that needs to be determined is what percentage of positrons annihilate in the kapton foil. This depends on the thickness of the foil as well as the sample material as samples with heavier nuclei cause more positrons to bounce back into the foil. A semi-empirical formula by Bertolaccini-Zappa formula 4 exists for this that can be used to estimate the intensity of this lifetime component ($I(\%)$) based on just the thickness (s) and density (ρ) of the foil and the (average) atomic number of the sample (Z) [11]. The kapton foil used in Proto1 was $8.00\mu m$ thick.

$$I(\%) = 0.905 * Z^{0.619} * (\rho s)^{\frac{1.49}{Z^{0.235}}}, \tag{4}$$

Positronium states last for a relatively long time, and consequently they cause a long lifetime component of 1500 ps. Long lifetime components can be hard to distinguish from the radioactive background, but we can use this to find the correction percentage for the positronium component. First, we measured the real background of the experiment using the same method as we did for finding the background N_{random} in Section 3.3.1. Then, we allow PALSfit to try and fit a background to the lifetime spectrum, but because of the long lifetime component of positronium, PALSfit thinks it is part of the background and incorrectly fits a higher background. We can then add a positronium source correction component to the program and change its correction percentage until the background fitted by PALSfit agrees with our background calculations.

Once we know the positronium and kapton source correction components as well as the lifetime of the sample itself, we only need to figure out the lifetime component of the 22Na salt itself. We know that 22Na salt has a positron lifetime of 400 ps. As this is the last unknown component, we can figure out its intensity by inputting it as both a source correction and a "sample lifetime" component into PALSfit. As we increase the intensity correction percentage of the 22Na source correction parameter, the intensity of the fake sample lifetime 22Na component decreases as the source correction is substrating it from the results. When the 22Na lifetime intensity vanishes, we know that we have the correct source correction intensity.

As the source correction intensities changed based on the sample used and how the source-sample-sandwich is constructed, they need to be re-evaluated on a per-experiment basis, but this same process can be repeated every time to get reliable results. With pure samples with known bulk lifetimes, obtaining source correction parameters can be very accurate, but the accuracy can drop substantially when measuring unknown samples with potentially multiple lifetime components.

4 Testing and optimization

4.1 Validating the source

Since a special type of kapton source, Proto1, had to be made for a new setup, it was necessary to demonstrate that this source can be used for positron lifetime measurements. In order to do this, lifetime measurements were run on a standard two detector setup using the new Proto1 source and a normal aluminum source (Al125). A total of three samples were measured with both sources, these samples were silicon, nickel, and germanium. The samples were new and hadn't been measured with any PALS equipment yet, though being known elements, there were literature values to indicate what kind of results could be expected. Lifetime spectra were fit into the resulting data using PALSfit software. The resulting lifetimes and source correction parameters used can be seen in table 1.

Al	Si		Ni		Ge	
TZ (chn)	248		246		249	
FWHM (ps)	285		271.8		285	
BKG (counts)	6.073		3.8613		4.3504	
Positronium (ps)	1500	0.57%	1500	1.13%	1500	0.62%
Sodium (ps)	400	3.63%	400	6.33%	400	3.91%
Aluminum (ps)	220	2.11%	220	4.63%	220	5.37%
Lifetime (ps)	224.9		122.8		233.6	
Proto1 (G)	Si		Ni		Ge	
TZ (chn)	250		247		250	
FWHM (ps)	282.1		276.5		285.9	
BKG (counts)	14.8978		9.9197		11.4015	
Positronium (ps)	1500	2.35%	1500	10.40%	1500	3.91%
Sodium (ps)	400	11.20%	400	28.92%	400	13.00%
Aluminum (ps)	385	5.13%	385	7.77%	385	8.41%
Lifetime (ps)	236		169.1		243.8	
Proto1 (F)	Si		Ni		Ge	
TZ (chn)	250		247		250	
FWHM (ps)	282.1		276.5		285.9	
BKG (counts)	14.8978		9.9197		11.4015	
Positronium (ps)	1500	2.24%	1500	9.96%	1500	3.84%
Sodium (ps)	400	16.83%	400	41.95%	400	17.58%
Aluminum (ps)	385	5.13%	385	7.77%	385	8.41%
Lifetime (ps)	224.9		122.8		234.7	

Table 1: Table showing the source correction parameters and measured lifetimes from both experiments. Aluminum source was only analyzed in PALSfit's "guessed" mode, while two analysis were made for proto1, one in "guessed" and another in "fixed" mode.

Since Proto1 was still a relatively untested source, two different fits were made.

The first one, "Proto1 (G)" with "guessing" the lifetime of the sample, like we would do with a hypothetical unknown sample, with the source correction parameters determined via the process described in section 3.3.2 with fixed values. For "Proto1 (F)", we fixed the lifetime to the results we measured with Al125 but let PALSfit guess and fit the intensities of the source correction parameters. This was done to have a point of comparison to see if source correction parameters would need to be altered dramatically to produce similar results as AL125, or if any changes in the results could be attributed to random chance and measurement errors.

From the results, we can see that the with Silicon and Germanium samples resulted in similar lifetimes between the Al125 source and Proto1. There seems to be a systematic error of about 10 ps, the cause of which is unclear, but it shouldn't matter for comparisons between elements and this systematic error could be accounted for if it is consistent with other elements as well. Previous experiments on pure silicon and germanium samples have obtained positron lifetimes of 218 ps and 228 ps respectively [12], which are even lower than the results with AL125, but show similar relative difference in positron lifetimes.

The results with the nickel sample, however, did not produce similar results at all, with a massive 56.3 ps difference in average lifetime between Proto1 (G) and Al125. The source correction intensities also needed to be adjusted to unusually high values to get even reasonable lifetime results, and in order to make Proto1's results match those of Al125, the intensity for the sodium lifetime component would need to be raised to above 40%, which would be an absurdly unbelievable result. Previous experiments with pure nickel samples have given positron lifetimes ranging from 95 ps [13] to 110 ps [14], which are in better agreement with measurements done with the aluminum source.

The most likely reason for the discrepancy in lifetime results is that the *sandwich* was poorly made for the experiment using the Proto1 source and nickel sample. If the *sandwich* wasn't tight enough, there would be more space between the source and the sample for positronium to form, which could explain the unusually high positronium component.

It is also possible that the nickel sample was not as pure as expected and gave multiple lifetime components and these additional components got incorrectly characterized into the source correction intensities in the data analysis process. This could explain the unusually high sodium correction component with the measurements using even AL125 (as the sodium component, is also affected by backscattering from the sample, it should be proportional to the atomic number of the sample, but instead it is higher with nickel than with germanium), and why even measurements with that source had more error compared to literature values than with germanium and silicon samples.

Regardless, since the silicon and germanium results agreed extremely well with each other and literature, while the results with nickel were strange, we chose to discard the nickel measurement results. Further testing would need to be done on these nickel samples to understand why it produced such bizarre lifetime components.

Ultimately, Proto1 was deemed to be a suitable positron source for PALS measurements with a noted 10 ps systematic error that might need to be accounted for

in future experiments using the source.

4.2 Energy calibration and time resolution.

The plastic scintillator detectors work by sending an electric pulse to the computer whenever it detects a photon and the height of this pulse is proportional to the energy of the detected photon. In order for the detectors to be used for PALS measurements, they need to be calibrated to detect photons of the right energy (511 keV for the stop detectors and 1.28 MeV for the start detector), and this is done by setting lower and/or upper thresholds so that only pulses of a certain height, corresponding to energy we want to detect, are counted. While in other types of PALS setups, it is possible to set an upper and lower threshold for energy detection, the TCSPC card in our setup could only allow us to set a lower threshold, meaning any photons above this threshold would always be detected and counted. (This would increase false counts in a 2 detector setup as the *stop* detector could count the higher energy 1.274 MeV photons as *stop* signals, but since the three detector setup requires the the detection of two photons in coincidence, this is not an issue.)

This threshold level needs to be set for each detector independently, but in order to do that, we need to find the relationship between the energy of the detected photon and the height (voltage) of the pulse sent by the detector after detecting that photon. These correlate, but each detector is slightly different, and the detection thresholds need to be tuned as precisely as possible (but accounting for the energy resolution of the detectors). In standard PALS setups, this can be done using a Multi Channel Analyzer (MCA), however our setup was not equipped with an MCA and thus a new innovative approach had to be taken to set these thresholds.

First, a ^{22}Na source was set between the detectors with the detection threshold set as low as possible and the detectors would count photons over a 5-minute period. Then the threshold would be raised slightly, and the experiment would be repeated again. With no lower threshold, the number of counts is massive as even random electronic noise is counted, but as the threshold level is increased, fewer photons are counted as they fall below the detection threshold. Eventually, as the threshold is high enough that 511 keV annihilation photons are no longer counted, the count rate would plummet. As the threshold is gradually raised, eventually even the 1.28 MeV *start* gammas would not be counted, and the count rate would plummet a second time. We can then plot the count rate as a function of the threshold level and since we know the sudden drops in the count rate correspond to specific energies, we can use this to find the relationship between detected photon energy and threshold level. This *threshold response spectrum* can be seen in figure 5.

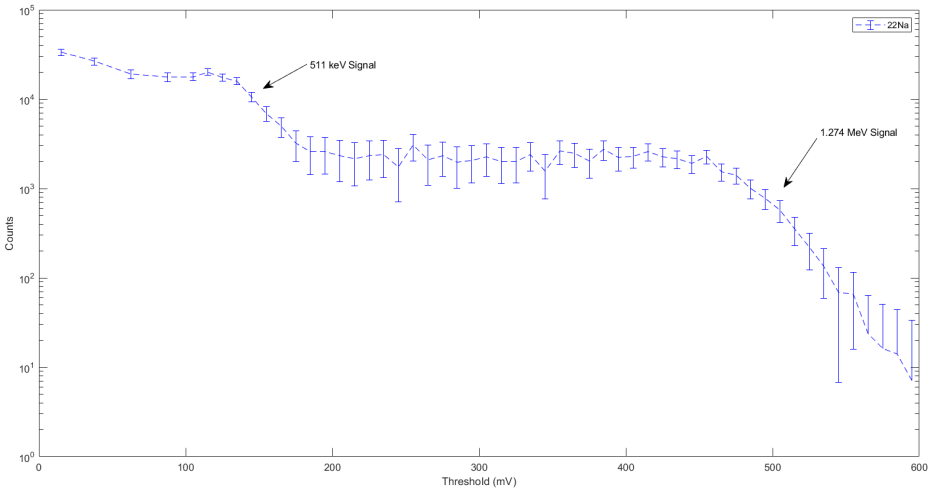

Figure 5: Reconstructed threshold response spectrum for 22Na for detector 0. The first drop in counts corresponds to the 511 keV *stop* signal, while the second drop corresponds to the 1.274 MeV *start* signal.

In addition to using ^{22}Na, this process was also repeated with ^{137}Cs and ^{133}Ba. This was done for two reasons. The first one was to confirm our results, the most common emission gammas from these two elements (661 keV for ^{137}Cs and 356 keV for ^{133}Ba) should have their own detection thresholds and the resulting threshold to photon energy relationship should agree with the results from the ^{22}Na experiment. If this were not the case, we would know that something would have gone wrong. The second reason was that both of these are fission products with common emission photons close to the energy of the 511 keV annihilation gammas. In particular, false counts from ^{133}Ba could be completely eliminated by setting the lower threshold below 511 keV, but above 356 keV. The resulting threshold response spectrums for ^{137}Cs and ^{133}Ba as well as potential detection thresholds can be seen in figure 6.

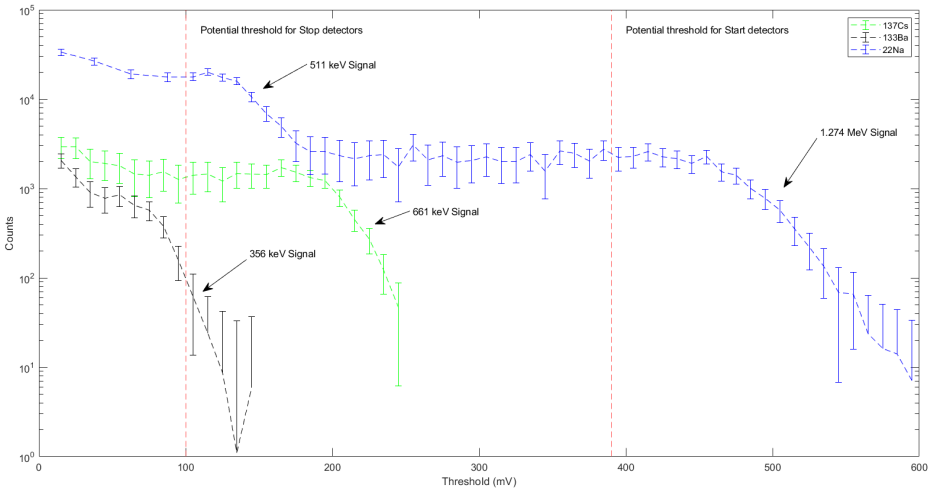

Figure 6: Threshold response spectrum for all three tested elements. Potential detection thresholds for the *stop* and *start* detectors are shown at 100 mV and 390 mV respectively. These are set slightly before the corresponding signals to account for poor energy resolution.

4.3 Testing the setup

To test the setup, we simulated the effect of measuring a radioactive sample by measuring a known non-radioactive sample, but placing several sources of gamma radiation in close proximity to the sandwich. This would allow us to have an expected sample spectrum from a regular low-activity PALS measurement to compare against, and we would know if the system was producing incorrect results.

The main experiment was done by placing ^{226}Ra samples near the sandwich and doing a lifetime measurement, then repeating this with no additional source as a control measurement. Both of these measurements were done in both double and triple coincidence modes, and the sample being measured was the same germanium sample used for source validation in section 4.1. Additionally, a shorter third measurement was done with ^{60}Co sources near the sandwich instead of ^{226}Ra sources. As this measurement was shorter, no direct comparisons could be made with the data, but by normalizing the resulting lifetime spectrum, this could be used to further demonstrate the expected distortion of the lifetime spectrum.

It is worth noting that all six measurements were done with using Proto1 as the positron source. As it is more powerful than the average source used in PALS experiments, it also contributes to additional radioactive background.

5 Results and conclusions

A germanium sample was measured in both double and triple coincidence modes using our finished setup. Following the methodology described in section 4.3, there was one experiment done with additional ^{226}Ra radiation sources near the sandwich, and another control experiment with no additional sources nearby. The results from these from these measurements can be seen in figure 7.

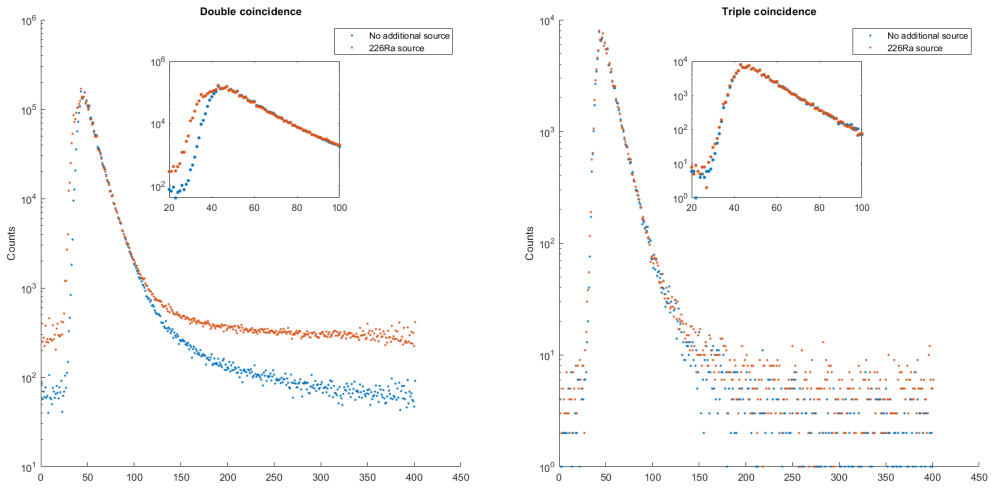

Figure 7: Double and triple coincidence measurement results with and without ^{226}Ra source.

In the case of the double coincidence measurement, clear distortion of the shape of the graph can be seen near the peak, with the presence of the 226Ra source causing additional counts to shift the peak to the left. Additionally, the increased background radiation is also distorting the tail of the graph.

In the triple coincidence measurement neither of these distortions are present; the peaks of both graphs align extremely well, and background radiation is almost entirely gone, with counts per bin consistently in single digits.

With a third experiment using ^{60}Co instead of ^{226}Ra as a source of additional radiation, similar results are found. In figure 8 the double coincidence spectrum distorts, while the triple coincidence shows similar graph shape regardless of additional radiation source, or lack thereof.

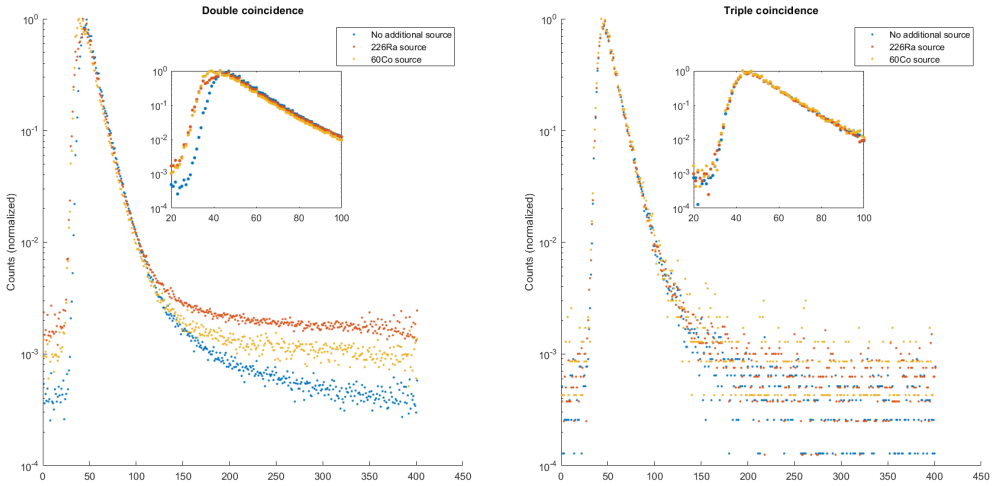

Figure 8: Double and triple coincidence results with ^{226}Ra, ^{60}Co and no additional source. Counts are normalized between 0 and 1, since the measurement with ^{60}Co was shorter.

The count rate in double coincidence mode was observed to be 20% higher when the ^{226}Ra source was present compared to the control measurement. In triple coincidence, the presence of the radium source only increased the count rate by 3%. This increase in count rate for triple coincidence mode is likely due to false events counted by the *start* detector. A significant portion of the photons emitted by ^{226}Ra are above the detection threshold set for the *start* detector, so some random emissions from radium are detected in coincidence with positron annihilation when ^{226}Ra and ^{22}Na decays happen in close proximity. This can be somewhat mitigated by increasing the detection threshold on the *start* detector at the cost of losing a portion of real counts as well due to poor energy resolution of the detectors.

The overall count rate was still significantly lower in triple coincidence compared to double coincidence. Even with no additional source present, the count rate from double coincidence was 20 times higher than with triple coincidence. In part this is due to significantly lower radioactive background in triple coincidence, but a significant proportion of real lifetime events go undetected by a three detector measurement, as the likelihood of successfully detecting three photons is significantly lower than only needing to detect two. This means that triple coincidence measurements need to be longer to achieve a suitably high number of counts compared to double coincidence measurements. Still, the count rate observed in our setup was high enough that this is just an inconvenience.

5.1 Following developments

Development and optimization of the three detector positron lifetime spectrometer continued after this initial testing, and was eventually named *PALSRaM* [5]. The

setup installed in the University of Helsinki and used for studying radioactive materials. Additionally, a copycat setup was installed in the hot laboratory of the European Joint Research Center in Karlsruhe, Germany and was used for the original purpose of studying nuclear fuel. This setup also included the glovebox and was mounted vertically on rails following the original concept.

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