

## Testing an Ortec Lifetime System

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**Keywords:** positron annihilation, lifetime, zirconia, silicon.

### Abstract

We report preliminary performance tests of an ORTEC PLS lifetime system based on plastic scintillators and analog electronic system. A variety of samples was measured, from metals (Cu, stainless steel), across semiconductors (Cz-grown silicon, ZnSe) to nanostructured ceramics (ZrO<sub>2</sub>). All results obtained are compatible with literature reports and indicate the lifetime resolution of the whole system as 180 ps.

### Introduction

With the advent of fast digital electronics [1] the use of analog lifetime systems could become questionable. On the other hand, the spread of existing experimental results and in some cases - like pure metals - their still poor agreement with the theory [2] make new measurements (and analysis packages [3]) still useful. Additionally, time resolution in lifetime beam experiments is hardly better than 250 ps [4, 5] so mutual testing of different techniques is needed in order to obtain a kind of normalization, at least for the most simple systems, like pure, defect-free metals or Si.

### Experiment

The ORTEC PLS system, as acquired in March 2010 is based on plastic scintillators (St. Gobain BC418) and RCA 8850 photomultipliers. The electronics consists of two 538B constant fraction discriminators, the 414A fast coincidence module and the 567 time-to-amplitude converter. The system is under testing in a rather unusual configuration, using a very low-intensity (0.5  $\mu$ Ci) <sup>22</sup>Na source in 7  $\mu$ m thick kapton sandwich, with one side covered by a stainless steel (AISI 316LN) 1 mm thick plate. Due to the weak source, the coincidence count was low, 2-3 counts/sec. We performed for each sample several runs, typically 10-20, with 12, 24 or 48 hours acquisition time. The back plate in our radioactive source acts as its mechanical reinforcement and simulates conditions of some background signal present for example in beam experiments. As shown later in this paper, the presence of the background sample made the analysis much more tedious than in usual configurations, i.e. with two identical samples on both sides of the radioactive source. This is mainly due to the changing contribution from backscattering for samples with different specific densities and/or atomic mass.

The analysis of lifetime spectra was performed with the LT package by J. Kansy [6]. In our previous paper [4] we have tested extensively that package vs. the genetic algorithms [7] and the POSFIT software. The LT package is quite flexible, in particular it allows the use of the trapping model apart from a straightforward multicomponent analysis.

### Results

We have performed tests on a number of samples belonging to different kinds of materials. The choice of the samples aimed to test the ability of the combined method (Ortec hardware and LT

software) to detect both short lifetimes and few components, like in silicon, as well as intermediate lifetime with poorly separated components, like in graphite. We started the tests from a reference stainless steel sample. AISI 316LN stainless steel is an iron based non-corrosive, non magnetic steel with low (0.08%) carbon contents and high chromium and nickel contents (typically 16% and 13%, respectively). The two main alloy components, together with 2% of Mo allow to maintain the austenite crystallographic structure of the steel after cooling. Our samples come from cold rolling process, with no annealing afterwards, with dislocations being the dominant defects. An expectation would predict the lifetime in the stainless steel somewhat close to the pure iron or chromium, which is the main component (20%) on that steel. The theoretical values for both elements, Fe and Cr are identical - 101 ps. In extensive measurements in Trento laboratory [9] two component lifetime spectra were obtained,  $111 \pm 1$  ps and  $257 \pm 10$  ps ( $8.6 \pm 1\%$ ) for iron and  $83 \pm 1$  ps and  $248 \pm 2$  ps ( $45 \pm 1\%$ ) for chromium. In our measurements of 316LN steel none of these components appeared.

Table 1. Lifetime analysis for AISI 316LN samples in three different experimental runs.

Run No.	$\tau$ [ps]	variance
1	145	1.05
2	146	1.04
3	147	1.05

Instead of lifetimes near to 100 ps, we observed in the stainless steel a clearly dominating lifetime component of 146 ps, see Table 1. However we note the only moderately good variance of the fit (never better than 1.05) which indicates the presence of at least another lifetime component in the spectra - we are not able to separate this component in our system. The 146 ps lifetime probably comes from trapping positrons in tetrahedral voids inside the austenitic structure and/or in defects induced by cold rolling. A similar value (145-150 ps) was declared by Kawaguchi and Shirai [8] as the defect lifetime in the same type steel. Those authors studied the lifetime components in the steel subject to strain cycles and concluded that the above cited defect lifetime does not vary in the strain cycles, in contrast to the "matrix" lifetime which lowers with the strain cycles from about 100 ps to as little as 40 ps. After prolonged strain cycles the intensity of the defect life time rises to as much as 80-90%. We were not able to detect such short lifetimes as Kawaguchi and Shirai, but the variance value indicates its probable presence. In all subsequent measurements the lifetime for the back plate was kept fixed at 146 ps and about  $50 \pm 5\%$  intensity. The source component is discussed below, with the example of Si samples.

### Copper samples

Measurements from Trento [9] yielded for the Cu annealed monocrystals a single lifetime of 115 ps, in reasonable good accord with the theory (106 ps [10] and 123 ps [2]) and other experiments (110 ps [11] and 122 ps). Copper samples for the present measurements were oxygen-free type, but with only commercial purity and with no annealing. Nevertheless, the 115 ps lifetime component was clearly separable in all spectra and the obtained variance resulted very good (1.01). The second lifetime component is about  $240 \pm 10$  ps, with about 27% intensity [12].

### Silicon samples

Positron lifetimes in Czochralski-type silicon were extensively studied in our previous papers [13]. Commercially available "as-grown" Czochralski-type silicon wafers usually undergo a thermal treatment for 20 hours at 450 °C just after the growth from melt. This technical procedure known on the phenomenological basis from the very beginning of the silicon era aims to remove the electrical donor activity of oxygen atoms (which in Cz-silicon are in about  $10^{18}$  cm<sup>-3</sup> concentration). The treatment does not remove oxygen atoms from silicon but dissolves them into interstitial positions. Our measurements [13] have shown that this treatment brings the positron lifetime in silicon down to about 221 ps, while for the true "as-grown" silicon this lifetime is somewhat higher (223-225 ps) and/or shows additional longer components. Thermal treatments at higher temperatures, and in

particular with the use a high pressures, lead to formation of oxygen precipitates and/or silica non-stoichiometric nanoclusters [14]. In lifetime spectra [13] additional, long components ( $500 \pm 100$  ps) can be then separated. The theory [15] attributes such long lifetimes to nanovoids in silicon crystalline structure. Present measurements were performed for p-type Cz-grown samples ( $5 \Omega\text{cm}$ ) obtained from 5'' wafers, cut and mechanically polished but did not underwent any thermal treatment after the growth from melt. Apart from the lifetime of 220 ps, typical for the silicon bulk, we observed a second lifetime component, of about  $450 \pm 10$  ps and  $24 \pm 2\%$  intensity. The variance of the fit was surprisingly good,  $\chi^2=1.02$  and the residuals too (see Fig. 1).

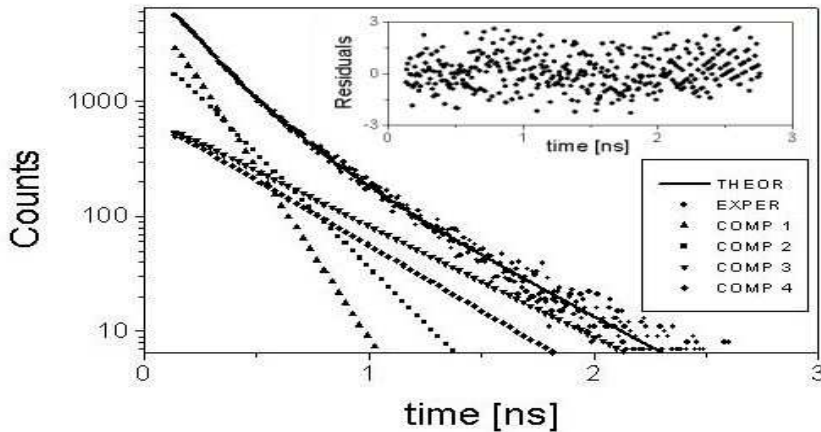


Fig. 1. The residuals for silicon Cz-grown sample.

annealing at  $1200 \text{ }^\circ\text{C}$  (see Fig. 4 in [16]). In our samples this component is probably due to mechanical processing of the silicon wafers after the growth.

#### Source and back-plate contributions

Promising results obtained with silicon allowed us to study in detail the contribution of the source, of the back-plate and the resolution of the system. The study of the variance vs. the contribution from the radioactive source contribution, i.e. the single 382 ps component due to kapton sandwich, is shown in Fig. 2. Minimum for the variance in the three component analysis (146 ps, 45% fixed for the stainless steel, and two components for silicon) is observed for 12% source contribution, in good agreement with our previous analysis for Cz-grown silicon [13]. The analysis was performed assuming the time resolution of the system of 180 ps. The study of the variance dependence vs. the time resolution is shown in Figure 3. Minimum of the variance is observed at 180 ps resolution. This is somewhat worse than the resolution of 148 ps declared by the manufacturer for  $^{60}\text{Co}$  source and narrow windows but much better than the 230 ps value declared by the manufacturer for  $^{22}\text{Na}$  source and broad start and stop windows of the signal discriminator. Obviously, the above analysis is far from being complete. In the first instance the trapping model should be used in cases like silicon with mechanical defects. However, the use of trapping model is somewhat difficult in our present experimental configuration. We perform the analysis treating the lifetime value and the intensity in the background plate as fixed values. In the trapping model, this contribution from the back plate would be mixed up with samples lifetimes.

In a trial for silicon, the trapping model would allow to lower the variance of the fit to as little as 1.0002 but then the back-plate contribution would be 60%. Fixing the back-plate contribution to 50% worsens the variance to 1.18. In general, the back-plate contribution is the main source of uncertainties in our analysis. The data presented in Fig. 2 and 3 have been obtained fixing its contribution to 45%, as can be expected from our previous studies of the source contribution vs. the atomic number  $Z$  in pure elements done for the ref. [9] but remained unpublished.

Trials of changing the back-plate contribution in the range 40-50% worsen the variance of the fit. A dependence with the longer lifetime component in silicon was observed: the same variance can be

obtained for the 50% back-plate contribution as for the 45% contribution but then the  $\tau_2$  in silicon rises than from 440 ps to 500 ps.

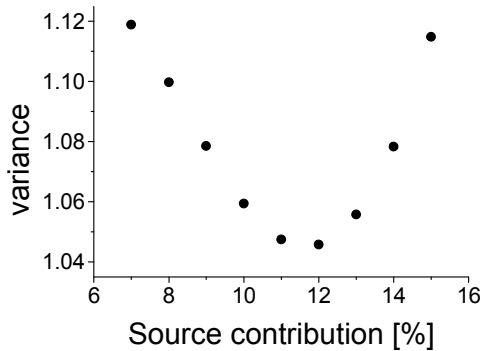


Fig. 2. Studies of the source component for lifetime analysis in Cz-grown samples.

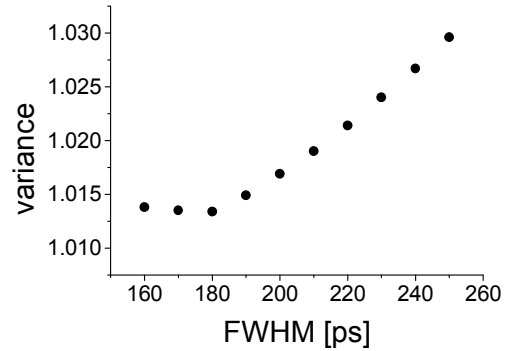


Fig. 3. Studies of the time resolution of ORTEC system using Cz-grown Si samples.

### Pyrolytic graphite

It has numerous technical applications from electrodes in spark discharges to neutron moderators in nuclear reactors. Highly oriented pyrolytic graphite is a promising material for special purpose electronics. In present measurements we used low-cost pyrolytic graphite showing - thanks to its low density - a diamagnetic levitation in strong magnetic fields. The material shows macroscopic irregularities and internal porosity. For our samples of pyrolytic graphite three lifetimes were found, 208 ps (34%) 233 ps (20%) and 420 ps (46%).

Table 2. Three component analysis of positron lifetime in pyrolytic graphite.

Run No.	$\tau_1$ [ps]	$\tau_2$ [ps]	$I_2$ [%]	$\tau_3$ [ps]	$I_3$ [%]	variance
1	208	233	20.2	420	46.0	1.01
2	208	233	18.2	420	50.4	1.01
3	208	233	21.8	420	50.7	1.01

Two components (208 ps and 233 ps) were observed also in highly-oriented pyrolytic graphite and attributed to annihilation in bulk and in single vacancies [17]. The third of the presently measured components comes probably from annihilation in some bigger voids in a rather loose structure of present samples. More detailed studies, with some microscopic techniques would be needed.

### Hydrothermally grown zirconia

Yttrium-stabilized zirconia finds applications in imitations of diamond; nanoporous zirconia is used in gas sensors and as a membrane in high-temperature fuel cells. Present zirconia samples were obtained by hydrothermal microwave driven process. Details of the sample preparation process are described in our previous paper [18]. In that study, using a positron beam, we found that hydrothermally grown  $ZrO_2$  samples show some significant porosity extending into the depth of several hundreds nm. Annealing at high temperatures, and in particular in oxygen atmosphere changes the crystalline structure and also the porosity. In the present study we examined three samples, annealed at 700 °C, 800 °C and 900 °C. Results are shown in Table 3.

Prohazka et al. studied positron lifetimes in pure zirconia powder, yttrium stabilized powder, sintered and in crystallized samples. In pure zirconia powder they found four lifetime components,  $\tau_1=189$  ps and  $\tau_2=373$  ps with 45% intensities each and  $\tau_3=2$  ns and  $\tau_4=34$  ns with 1.5% and 7.5% intensities, respectively. The analysis of our samples shows only two components,  $\tau_1=180$  ps and  $\tau_2=360\pm 10$  ps. The intensity of the second component rises rapidly from about 20% to 70% with the change of the annealing temperature from 700 °C to 900 °C.

Table 3. Positron lifetimes in hydrothermally grown zirconia.

Run No.	$\tau_1$ [ps]	$\tau_2$ [ps]	$I_2$ [%]	variance	T [°C]
1	180	370	19.8	1.01	700
2	180	361	76.4	1.03	800
3	180	352	70.6	1.02	900

## Conclusions

Present tests of our analog Ortec lifetime system has been performed with a very weak radioactive source and using only one sample, together with a reference, "back" sample made of stainless steel. In spite of such a configuration the system allowed to measure a variety of materials. Using LT analysis programme we were able to distinguish both short lifetimes, like 115 ps in copper and several close lifetime components, like in pyrolytic graphite. Good variance values were obtained for all samples, indicating consistently the lifetime resolution of the apparatus of about 180 ps. Main backwards of the present research remain the relatively low statistics of spectra, making plausible an overestimation of the fit quality (the variance values very close to 1.0) and the presence of the background plate (an additional lifetime present in the spectra). A new radioactive source is under delivery what will allow avoiding these problems. In any case, some reference samples, interchangeable between different laboratories would be also needed.

## Acknowledgements

The ORTEC PLS system has been purchased within the NLTK laboratories (grant no. POIG 02.02.00-00-003/08) at NCU in Torun. This work has been partially supported by the Ministry of Science and Higher Education (Poland) by Grant No. N N202 128839 for the years 2010-2012.

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