Tomographic Positron Annihilation Lifetime Spectroscopy

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Abstract. Positron annihilation lifetime spectroscopy serves as a perfect tool for studies of
open-volume defects in solid materials such as vacancies, vacancy agglomerates, and
dislocations. Moreover, structures in porous media can be investigated ranging from 0.3 nm to
30 nm employing the variation of the Positronium lifetime with the pore size. While lifetime
measurements close to the material’s surface can be performed at positron-beam installations
bulk materials, fluids, bio-materials or composite structures cannot or only destructively
accessed by positron beams. Targeting those problems, a new method of non-destructive
positron annihilation lifetime spectroscopy has been developed which features even a 3-
dimensional tomographic reconstruction of the spatial lifetime distribution. A beam of intense
treißstrahlung is provided by the superconducting electron linear accelerator ELBE (Electron
Linear Accelerator with high Brilliance and low Emittance) at Helmholtz-Zentrum Dresden-
Rossendorf. Since the generation of bremsstrahlung and the transport to the sample preserves
the sharp timing of the electron beam, positrons generated inside the entire sample volume by
pair production feature a sharp start time stamp for lifetime studies. In addition to the existing
technique of in-situ production of positrons inside large (cm³) bulk samples using high-energy
photons up to 16 MeV from bremsstrahlung production, granular position-sensitive photon
detectors have been employed. The detector system will be described and results for
experiments using samples with increasing complexity will be presented. The Lu₂SiO₅:Ce
scintillation crystals allow resolving the total energy to 5.1 % (root-mean-square, RMS) and
the annihilation lifetime to 225 ps (RMS). 3-dimensional annihilation lifetime maps have been
created in an offline-analysis employing well-known techniques from PET.

1. Introduction
Positron Annihilation Lifetime Spectroscopy (PALS) allows for studies of crystal lattice defects on the
nanometer scale and at low defect concentrations, open volumes in polymers, porosity and others.
Several techniques have been developed over the last decades which make use of either kinematical
observables of the annihilation radiation of positrons with electrons from the sample materials and of
the annihilation lifetime of positrons after injection into the sample material. Especially, long positron
annihilation lifetimes beyond a few ns are caused by voids and porous structures inside the sample
which may be of high relevance for material failure and/or structural failure. In the following we will
discuss a new method which allows correlating positron annihilation lifetime studies (PALS) with a
three-dimensional tomographic analysis of a bulk sample. The well-established PALS technique
makes use of radioactive positron sources like $^{22}$Na where the positron emission is accompanied by electromagnetic transitions from excited states in the daughter nuclei. Because photon emission is prompt with respect to the positron emission the time resolution of the additional photon detector adds to the overall achieved accuracy in lifetime measurements. This additional contribution can in turn be avoided using an accelerator-based source of positrons which furthermore offers the advantage of possible adjustments of the source strength and the time structure. We will describe in the following how a versatile source of positrons using a superconducting electron accelerator has been realized and used in position-resolved positron annihilation lifetime spectroscopy.

2. Positron production using a superconducting electron linear accelerator
The Helmholtz-Zentrum Dresden-Rossendorf operates the superconducting electron accelerator ELBE [1] which delivers energies up to 40 MeV and average beam currents up to 1.6 mA. It acts as a source of secondary radiation like coherent infrared light from free-electron lasers, THz radiation from undulators and dipole magnets, photo-neutrons produced inside a liquid-lead target, bremsstrahlung, and positrons. The layout of the facility and the various end stations are shown in Fig. 1. The superconducting technology allows adjustment of the electron beam time structure allowing versatile time-of-flight experiments (neutrons) or lifetime experiments as described below. The micro-pulse repetition rate can be selected as $2^n$ divisors of 26 MHz with $n = 0…8$. Typically, the micro-pulse repetition rate for positron annihilation lifetime experiments is chosen to be 26 MHz or 13 MHz with micro-pulse intervals of 38 ns or 77 ns, respectively. The micro-pulse width has been measured to be less than 5 ps using electro-optical sampling [2].

3. Tomographic Positron Annihilation Lifetime Spectroscopy
The gamma-induced positron annihilation lifetime facility [4, 5] has been extended by a set of position-sensitive photon detectors which will allow reconstructing a three-dimensional image of the distribution of positron lifetimes inside bulk samples [6]. Two pixelated photon detectors each made from $13 \times 13$ crystals of Lu$_2$SiO$_5$ of $4 \times 4 \times 20$ mm$^3$ volume [7] have been set. Each detector is equipped with 4 photomultiplier tubes and dedicated preamplifier and discriminator electronics have been developed in-house in order to achieve a timing resolution of 530 ps (FWHM) for the combined mean-time of all channels. The photon energy deposition is calculated using the sum of all four charge-integrated signals and individually calibrated crystal responses. The obtained energy resolution
is 11.4% (FWHM) at 511 keV photon energy for both detectors, respectively. Signal partitioning between the four photomultiplier tubes of one detector allows identification of the crystal in which the photon has interacted. Figure 2 shows a sketch of the setup with both detectors fixed perpendicular to the direction of the incoming photon beam.

Figure 2. Sketch of the positron annihilation lifetime tomography setup. The bremsstrahlung beam hits the sample which is mounted on a rotational stage allowing for 3D image reconstruction. Two pixelated Lu$_2$SiO$_5$ detectors detect both annihilation quanta in coincidence.

In the case of two single-pixel events between the two detectors the line-of-response is calculated as the 3-dimensional connection between the two pixel positions. The simplest image reconstruction is performed for a two-dimensional distribution which is reconstructed by calculating the point of intersection between the line-of-response and a plane centred in between both detectors.

The system has been tested with a two-dimensional structure made from Si/SiO$_2$ which had not been rotated during the experiment. Equal-sized pieces of monocrystalline Silicon and microscope slides of dimensions $12.5 \times 25 \times 0.8$ mm$^3$, see Fig. 3, have been fixed in between two thin-walled Kapton sheets and mounted parallel to the beam direction. Fig. 4 shows the projected images derived from correlated events between both detectors. The annihilation lifetime has been derived from the time difference between the mean time of all 8 photomultiplier signals and the radio-frequency of the accelerator.

Figure 3. 2D target consisting of Si wafer material (dark) and microscopy slides (light) wrapped into Kapton foils.

Figure 4. Two-dimensional distribution of two-photon annihilation integrated over all positron annihilation lifetimes (left) and the ratio of intensities gated for annihilation lifetimes in excess of 225 ps ($1\sigma$) by all lifetimes (right).

The lifetime-integrated distribution shows no distinct features except an enhancement in the centre of the image due to the increased solid angle for correlated detection. Gating on positron lifetimes in excess of 225 ps ($1\sigma$ of the timing resolution) clearly discriminates for regions inside the sample with enhanced formation of o-Ps, namely SiO$_2$. Both materials have been selected in order not to emphasize areas with different pair production yields. The timing resolution has not been corrected for the
inaccuracy of time-zero in the direction of the beam (x-axis in Fig. 4) which amounts to about 48 ps (RMS). Taking the mean timing of both detectors as the annihilation lifetime the inaccuracy due to different positron production sites perpendicular to the beam cancels. From Fig. 4 a lateral position resolution of 2.8 mm has been obtained by fitting one of the boundaries with the Gauss error function. Two three-dimensionally structured examples have been employed for further studies. One sample consists of a 25 mm diameter PTFE (Teflon) cylinder with embedded slabs made from Copper, Iron, and Aluminium having the same volume (12 mm$^2 \times 25$ mm) but different geometrical shapes, see Fig. 5 a).

Figure 5. Two of the samples studied using tomographic positron annihilation lifetime spectroscopy. Picture a) shows a PTFE cylinder of 2.5 cm diameter with inserts made of Al, Cu, and Fe with different shapes. Picture b) shows a cut-out part of a high-field magnet coil which is composed of copper wires and reinforcing materials consisting of PTFE (Teflon) and Poly(p-phenylen-2,6-benzobisoxazol) (Zylon$^@$). Lines indicate the planes of intersection used in the analysis.

A second sample has been cut from a high-field magnet coil resembling a 3-D structure composed of copper wires, reinforcing Zylon$^@$, and again Teflon, see Fig. 5 b). The structure is manufactured with the aim to withstand phases of high mechanical pressure during ms-long periods where magnetic field strengths of up to 91.4 T are generated inside the coils with peak currents reaching 100 kA from a capacitor bank with 50 MJ stored energy [8].

Figure 6. Reconstructed distribution of annihilation events for different regions of annihilation lifetimes after thirty iterations of the MLEM algorithm. Left picture shows all measured annihilations while the central picture is gated on a time interval between –1 ns to 2 ns (prompt). The right picture shows the ratio between prompt and all events discriminating for short annihilation lifetimes. The boundaries of the sample materials are indicated as thin lines.

In order to obtain a reconstructed distribution of annihilation events, the Maximum Likelihood Expectation Maximization [9] (MLEM) has been employed. The algorithm allows reconstructing iteratively a 3-D distribution of events from the set of all measured annihilation and the system matrix which comprises all possible combinations of detector pixels (169 $\times$ 169), rotation angles (180), image voxels (30 $\times$ 30 $\times$ 30) resulting in an overall size of 138 billion matrix cells. Two reconstructed
distributions which are obtained from full (Fig. 6, left) and gated on only prompt annihilation events (Fig. 6, center) are again used to generate a contrast enhanced annihilation-lifetime sensitive source distribution shown in Fig. 6, right side.

As the last case shown here, we study with the same procedure as described above the “real-world” sample shown in Fig. 5 b). While having a much more complicated internal structure and a variety of different materials with significant differences in positron annihilation lifetimes the procedure as described above still delivers contrast-enhanced images of the annihilation lifetime distributions after convergence of the MLEM algorithm. Fig. 7 shows cuts in the z, y, and x-planes. Here, the ratio between the prompt and the full lifetime spectrum reveals structures with rather long positron annihilation lifetimes indicating open (porous) regions of the structure.

Figure 7. Reconstructed distribution of annihilation events for different regions of annihilation lifetimes after convergence of the MLEM algorithm for the sample shown in Fig. 5 b). The three columns represent different intersections of the three-dimensional reconstruction. The three rows are gates on the integral lifetime (top row), gated on prompt emission (middle row), and the ratio between prompt and all events (lower row).

As the last setup in the analysis a complementary approach for a more quantitative analysis can be done which aims at determining the positron annihilation lifetime distribution for a selected voxel. Lifetime distributions for all voxels (here 30³) have been created by summing-up all list-mode events weighted by the intersection probability of each line-of-response for all rotational angles measured. Two selected lifetime distributions obtained in this way are shown in Fig. 7. It should be noted that also contributions from all voxels along the line-of-response contribute to the lifetime distribution; it is only that for a given voxel the statistical weight is enhanced.

Both distributions show a long lifetime component with a component of about 1.5 ns lifetime constant which is common for materials with significant positronium production such as polymers or porous structures. Clearly, both distributions show a significantly different intensity of the long lifetime component.
Figure 8. Positron annihilation lifetime distribution for two selected voxels from the reconstruction shown in Fig. 7. One of the distributions is parameterized by an exponential decay with 1.48 ns lifetime.

The newly developed system complements earlier developments of a positron annihilation microprobe which enabled high-resolution defect analysis at surfaces [10] or a high-energy positron beam for PALS studies in bulk material [11]. Further developments of the presented system aim at improving the position resolution by using smaller scintillator crystals and improved timing resolution by employing digital silicon photomultiplier readout.

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