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Position-resolved Positron Annihilation Lifetime Spectroscopy

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Abstract. A new method which allows for position-resolved positron lifetime spectroscopy studies in extended volume samples is presented. In addition to the existing technique of in-situ production of positrons inside large (cm^3) bulk samples using high-energy photons up to 16 MeV from bremsstrahlung production, granular position-sensitive photon detectors have been employed. A beam of intense bremsstrahlung is provided by the superconducting electron linear accelerator ELBE (Electron Linear Accelerator with high Brilliance and low Emittance) which delivers electron bunches of less than 10 ps temporal width and an adjustable bunch separation of multiples of 38 ns, average beam currents of 1 mA, and energies up to 40 MeV. 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 positron annihilation lifetime studies with high timing resolutions and high signal to background ratios due to the coincident detection of two annihilation photons. Two commercially available detectors from a high-resolution medial positron-emission tomography system are being employed with 169 individual Lu_2SiO_5:Ce scintillation crystals, each. In first experiments, a positron-lifetime gated image of a planar Si/SiO_2 (pieces of 12.5 mm × 25 mm size) sample and a 3-D structured metal in Teflon target could be obtained proving the feasibility of a three dimensional lifetime-gated tomographic system.

Introduction

Positron Annihilation Lifetime Spectroscopy (PALS) is a powerful tool which allows for studies of crystal lattice defects on the nanometer scale and at low defect concentrations, open volumes in polymers, porosity and others. Especially, lattice defects like dislocations, mono vacancies and vacancy clusters, as well as open-volume cavities are perfectly suited for investigations using positrons. 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, information about the electron density at the annihilation site is generated through Positron Annihilation Lifetime Spectroscopy (PALS) which correlates the distribution of annihilation lifetimes with the concentration and the type of defects.

Widespread standard techniques make 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 chapters, how a versatile source of positrons using a superconducting electron accelerator has been realized and used in position-resolved positron annihilation lifetime spectroscopy.

The accelerator facility ELBE

Starting shortly after the dawn of the new millennium a superconducting electron accelerator at the Helmholtz-Zentrum Dresden-Rossendorf came online [1]. Being the result of a development based on superconducting accelerating structures in the scope of the Tesla collaboration [2] a powerful new electron accelerator has been constructed which currently offers beam currents of up to 1.6 mA at maximum beam energies of 40 MeV. The facility is named Electron Linear Accelerator with high Brilliance and low Emittance (ELBE) and its high-intensity electron beam is being used as a driver for various types of secondary radiation e.g. coherent infrared light, THz radiation, photo-neutrons, bremsstrahlung, and positrons. A layout of the facility and the various end stations are shown in Fig. 1. Employing superconducting technology allows easy adjustment of the electron beam time structure which is crucial for time-of-flight experiments or lifetime experiments as described here. 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.

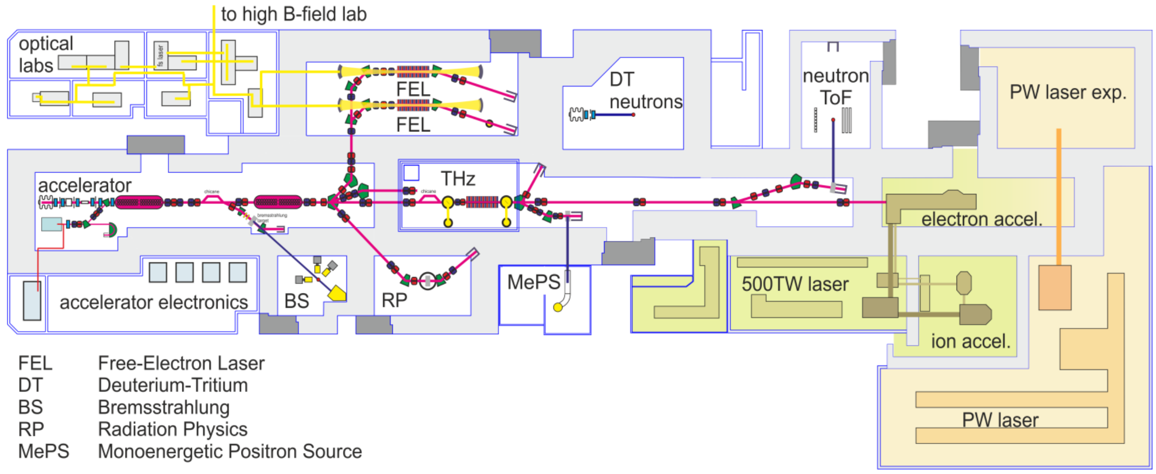

Figure 1. Layout of the ELBE centre for high power radiation sources. The overall width is about 100 m. Experiments discussed here took place at the bremsstrahlung facility labelled BS.

Position-resolved Positron Annihilation Lifetime Spectroscopy

The gamma-induced positron annihilation lifetime facility [3,4] 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. Two pixelated photon detectors each made from 13 × 13 crystals of Lu_2SiO_5 of 4 × 4 × 20 mm^3 volume [5] have been set up at the facility. Figure 2 shows a sketch of the setup with both detectors fixed perpendicular to the incoming photon beam. Each detector is equipped with 4 photomultiplier tubes. Dedicated preamplifier and discriminator electronics have been developed in-house in order to achieve a high time resolution of 450 ps per detector. The photon energy deposition is calculated using the sum of all four charge-integrated

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signals and individually calibrated crystal responses. The obtained energy resolution is 12.1% (FWHM) and 13.3% (IFWHM) 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.

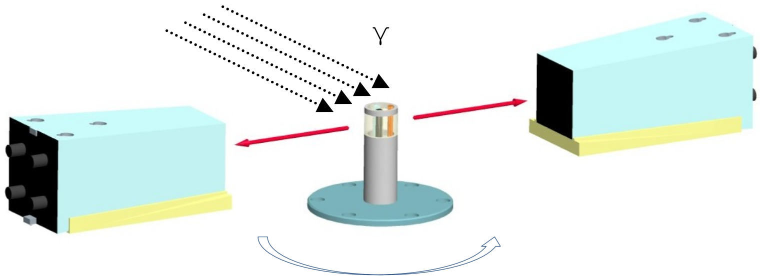

Figure 2. Sketch of the positron annihilation lifetime tomography setup. The bremsstrahlung beam hits the sample which is mounted on a rotational stage allowing for 3D image reconstruction. Two pixelated Lu_2SiO_5 detectors detect both annihilation quanta in coincidence.

The new system has been successfully 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.

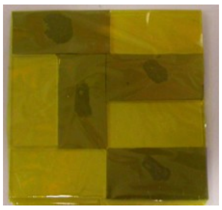

Figure 3. 2D target consisting of Si wafer material (dark) and microscopy slides (light) wrapped into Kapton foils.

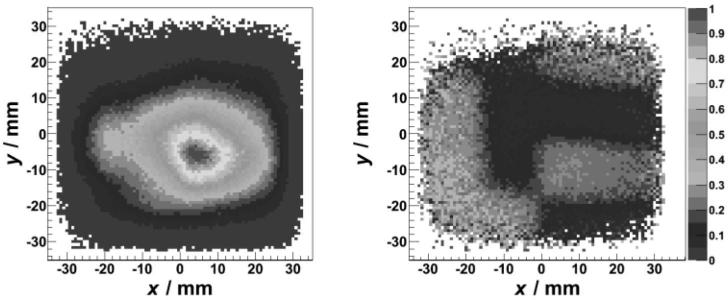

Figure 4. Two-dimensional distribution of two-photon annihilation integrated over all positron annihilation lifetimes (left) and the ratio of intensities gated for annihilation lifetimes in excess of 220 ps (1 σ) by all lifetimes (right).

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 220 ps (1 σ 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). Perpendicular to the beam, taking the mean timing of both detectors the annihilation lifetime inaccuracy due to different positron production sites 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. Further improvement and the reduction of image artefacts will be subject of a future publication.
As a fully three-dimensional example, a sample has been constructed consisting of a 25 mm diameter Teflon cylinder with embedded slabs made from Copper, Iron, and Aluminium having the same volume (12 mm^2 × 25 mm) but different geometrical shapes, see Fig. 5.

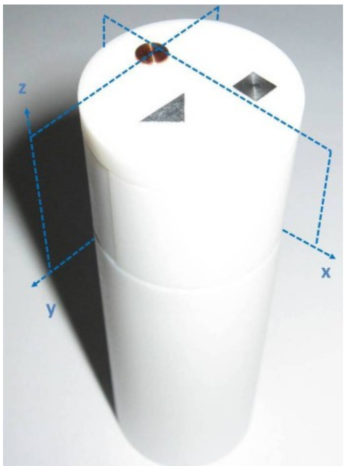

Figure 5. Three-dimensional sample made from Teflon with embedded slabs made from Cu, Fe, and Al. Dashed lines indicate the projections shown in Fig. 6.

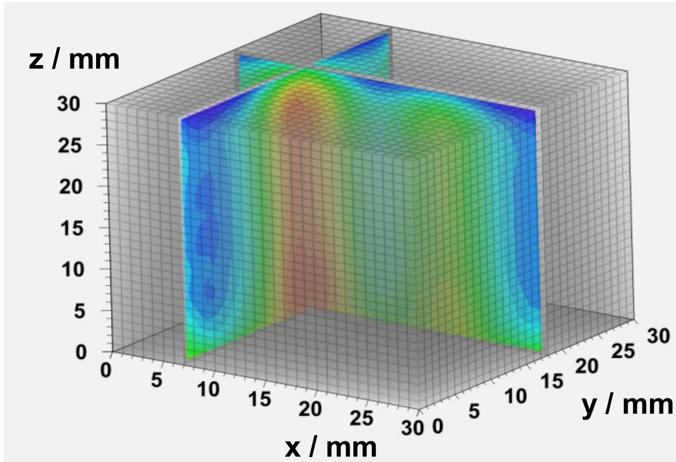

Figure 6. 3-D reconstruction of positron annihilation events gated on positron lifetime inside the sample shown in Fig. 5. Red colour indicates an enhancement of shorter annihilation lifetimes.

Figure 6 shows two perpendicular cuts through the 3-D reconstructed positron lifetime distribution. Clearly, the regions with shorter annihilation lifetimes can be identified. The newly developed system complements earlier developments of a positron annihilation microprobe which enabled high-resolution defect analysis at surfaces [6] or a high-energy positron beam for PALS studies in bulk material [7]. Further developments of the presented system aim at improving the position resolution by using smaller scintillator crystals and improved timing resolution.

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