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Positron-Annihilation Lifetime Spectroscopy using Electron Bremsstrahlung

A Wagner[1], W Anwand[1], M Butterling[1], T E Cowan[1,2], F Fiedler[1], F Fritz[1,2], M Kempe[1,2,†], R Krause-Rehberg[3]
[1] Helmholtz-Zentrum Dresden-Rossendorf, Institute of Radiation Physics, Bautzner Landstr. 400, 01328 Dresden, Germany
[2]Technische Universität Dresden, 01062 Dresden, Germany
[3]Martin-Luther-Universität Halle-Wittenberg, Institute of Physics, 06099 Halle, Germany
†deceased

E-mail: a.wagner@hzdr.de

Abstract. A new type of an intense source of positrons for materials research has been set up at the superconducting electron linear. The source employs hard X-rays from electron-bremsstrahlung production generating energetic electron-positron pairs inside the sample under investigation. CW-operation allows performing experiments with significantly reduced pile-up artefacts in the detectors compared to pulsed mode operation in conventional accelerators. The high-resolution timing of the accelerator with bunch lengths below 10 ps full width at half maximum (FWHM) allows positron annihilation lifetime spectroscopy (PALS) measurements with high time resolution. A single-component annihilation lifetime of Kapton has been measured as (381.3 ± 0.3) ps. Employing segmented detectors for the detection of both annihilation photons allows for the first time to perform a 4D tomographic reconstruction of the annihilation sites including the annihilation lifetime.

1. Introduction

Positron annihilation lifetime spectroscopy (PALS) serves as a suitable tool for the characterization of nanometer-sized lattice defects in crystals at low defect concentrations. Moreover, the production of Positronium allows studying open volumes in polymers and nanoscale porosities. Several complementary techniques have been developed in the past 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. 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. The new source is especially suited for extended bulk samples, or samples cannot be exposed to external sources of positrons (low-energy positron beams, radioactive sources), e.g. because they are imposing hazardous conditions (high pressure, high temperature, intrinsic radioactivity) to the source, or if the sample handling imposes difficulties (fluids, gases, organic samples). Here, we also report about a new method which allows correlating positron annihilation lifetime studies with a three-dimensional tomographic analysis of a bulk sample. The well-established PALS technique makes use of radioactive positron emitters (e.g. ^{22}Na) where the positron emission is accompanied by electromagnetic transitions from excited states in the daughter nuclei. Because photon emission happens promptly with respect to the positron emission (the lifetime of the 2^+ state in ^{22}Ne is only 3.6 ps) the time resolution of the additional photon detector adds to the overall achieved accuracy in lifetime measurements. Both, the contribution of the lifetime of the excited state and the contribution of the detector resolution can be avoided using an accelerator-based source of positrons. Furthermore, both the source strength and the time structure can be matched to the sample conditions under study. Here, we present a new versatile source of positrons using a superconducting electron accelerator for position-resolved positron annihilation lifetime spectroscopy.

2. The superconducting electron linear accelerator

The Helmholtz-Zentrum Dresden-Rossendorf operates the superconducting electron accelerator ELBE [1] which delivers electron beam energies up to 40 MeV and average beam currents up to 1.6 mA in continuous-wave mode. The electron beam serves 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, hard bremsstrahlung, and positrons. Due to individually accessible caves, the setup of experiments is possible while beams are delivered to other caves. The layout of the facility and the various end stations are shown in Fig. 1. The superconducting technology allows adjusting the time structure for time-of-flight experiments (neutrons), pump-probe experiments, or annihilation lifetime experiments as described in this contribution. Specifically, the micro-pulse repetition rate can be selected as $1/2^n \times 26$ MHz with $n = 0 \ldots 8$. 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, depending on the lifetime components expected in the sample under investigation. The micro-pulse width has been measured to be less than 5 ps using electro-optical sampling [2]. The ELBE installation operates as a 24/7 user facility while the free-of-charge access is granted through an external advisory committee.

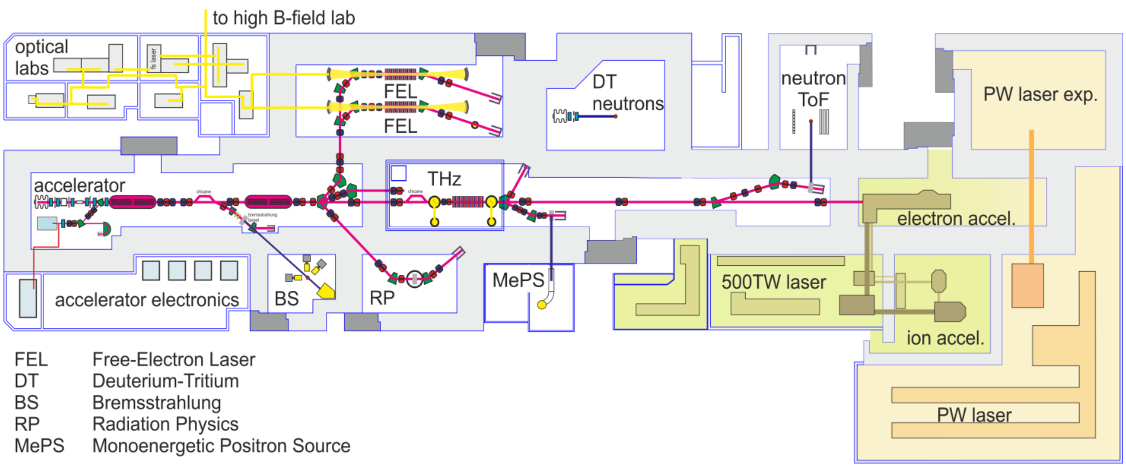

Figure 1. Layout of the ELBE centre for high power radiation sources. The overall width is about 100 m. Experiments discussed here took place at the bremsstrahlung facility labelled BS. Colour-shaded areas indicate high-power laser setups which are currently being developed for electron beam-laser interactions [3] and laser plasma ion acceleration.

3. Positron Annihilation Lifetime Spectroscopy of Bulk Samples

A new kind of positron annihilation lifetime spectroscopy setup has been realized at the bremsstrahlung facility [4] (see Fig. 2) at the ELBE LINAC. The pulsed electron beam with energies up to 16 MeV is converted into hard photons using thin Niobium radiator foils with thicknesses around 10^{-3} radiation lengths. Shaped by a 2600 mm long collimator the bremsstrahlung beam impinges onto

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a sample under study where it generates secondary positrons via pair production. These positrons thermalize in a few ps inside the sample. Characteristic annihilation quanta are being registered in coincidence by 4 sets of pairs of high-purity Germanium semiconductor detectors (HPGE) for accurate energy determination and BaF_2 scintillation detectors for accurate timing. Coincidence conditions and list mode storage of both energy and timing signals help efficiently reduce background. Furthermore, annihilation lifetime and electron momentum correlation studies, aka age-momentum correlation (AMOC) [5], can be performed with ease. Time walk effects of the timing discriminators can be minimized by offline corrections using the event-by-event recorded list mode data. In comparison to pioneering works at a normal conducting electron machine [6], the new installation makes use of continuous-wave (CW) beams which are only available at a superconducting (SC) accelerator. The CW–mode prevents detector pile-up effects which are present in macro-pulse bunched machines. Moreover, due to higher bunch charges the bunch lengths in normal conducting machines are significantly higher than in SC-machines. With typical ns-bunch widths positron annihilation lifetime measurements of metals become impossible.

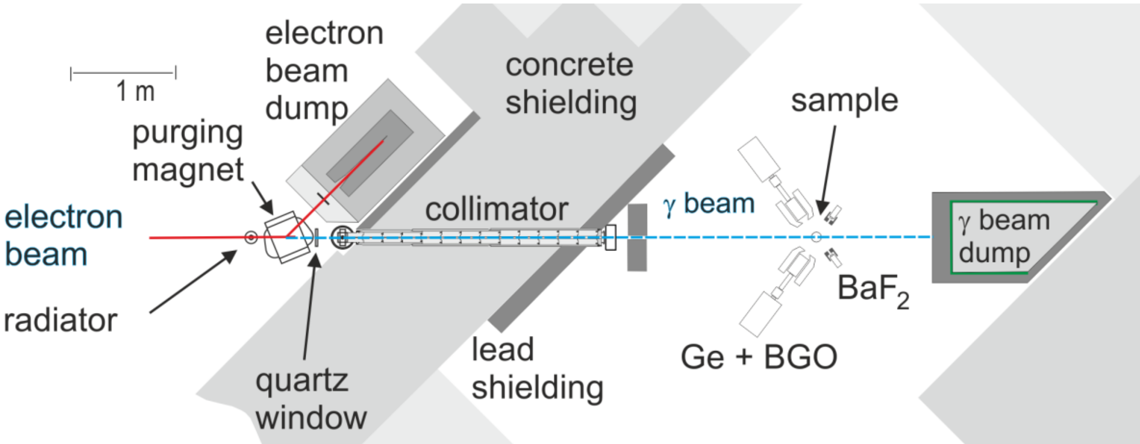

Figure 2. Layout of the bremsstrahlung facility at ELBE. The electron beam (red line) impinges from the left onto a radiator foil producing bremsstrahlung (blue line).

The facility has already been used in various positron lifetime studies and age-momentum studies (e.g [7, 8, 9]. As a benchmark example, we present here annihilation lifetime studies for bulk Kapton© (DuPont, Wilmington, NC), a polyimide widely used as cover material for radioactive sources in annihilation lifetime studies. Since positrons annihilate not only inside the sample material, the contribution inside the cover material has to be subtracted from the spectrum of the investigated sample material. Therefore, the source correction requires exact knowledge of the positron lifetime of the material. In contrast to other foil materials like Mylar, the annihilation parameters of Kapton© are not sensitive to temperature, making it an excellent candidate for PALS measurements. Up to now, there is a discussion about the positron lifetimes of Kapton©. The presence of just a single component with a lifetime of 382 ps was accepted for long time (for example [10]). On the other hand, a bimodal distribution with two components (280 ps with 30 % intensity and 410 ps) was reported in [11]. In order to help clarifying the discussing a measurement of the positron annihilation lifetime in Kapton© has been performed with positrons from pair production. The bulk Kapton© sample had dimensions of 7x7x7 mm^3. The beam repetition rate has been selected to be 77 ns in order to avoid pile-up effects from annihilation events with long lifetimes, e.g. from ortho-Positronium formation. From the combined analysis of the four independent lifetime measurements (see Fig. 3) one derives a lifetime of (381.3 ± 0.3) ps. Parameterizations with more than one lifetime component resulted in a worse χ^2 value. In order to check for the significance of Positronium formation at the Kapton© surfaces, a stack of foils has been studied, as well, but no indication for an extended lifetime component has been found. Fig. 3 shows the annihilation lifetime distributions measured by one of the four HPGe/BaF_2 combinations and the result of a parameterization with a single-component exponential and a constant value for beam uncorrelated background events. The structure observed at about 8 ns stems from background by scattered photons behind the sample.

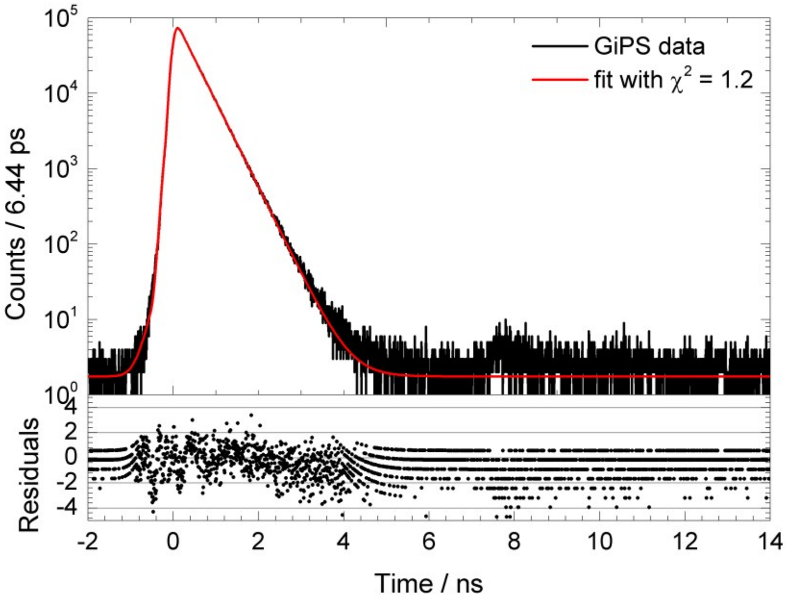

Figure 3. Positron annihilation lifetime distribution of bulk Kapton© for one of the four sets of HPGe/BaF2 detectors and result of the single-component exponential fit.

4. Tomographic Positron Annihilation Lifetime Spectroscopy

The positron annihilation lifetime facility discussed before has been extended by a set of position-sensitive photon detectors with the aim to reconstruct a three-dimensional image of the distribution of positron lifetimes inside bulk samples [3]. Two pixelated photon detector modules, each made from 13 × 13 crystals of Lu_2SiO_5:Y (LYSO) of 4 × 4 × 20 mm^3 volume (Siemens AG, Munich: Biograph® PET scanner), have been used. Each module is equipped with 4 photomultiplier tubes. Dedicated preamplifiers and timing discriminators have been developed resulting in a time 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 interacted. Figure 4 shows a sketch of the setup with both detectors fixed perpendicular to the direction of the incoming photon beam and rotational stage with the mounted sample. A three-dimensionally structured sample has been employed. It 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 × 25 mm) but different geometrical shapes, see Fig. 4. During the course of the measurement, the sample is rotated by 360° in steps of 2°.

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 three-dimensional distributions by overlaying all lines-of-response while rotating the sample around an axis perpendicular to the beam. Gating on positron lifetimes in excess of 225 ps (1 σ of the time resolution) discriminates for regions inside the sample with enhanced formation of o-Ps,

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namely regions with increased porosity (here: PTFE). 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 previous studies [3] a lateral position resolution of 2.8 mm has been obtained.

Figure 4. Left side shows the setup of two pixelated LYSO photon detectors employed in the tomographic system. Right side shows a PTFE cylinder of 2.5 cm diameter with inserts made of Al (triangle), Cu (disk), and Fe (square).

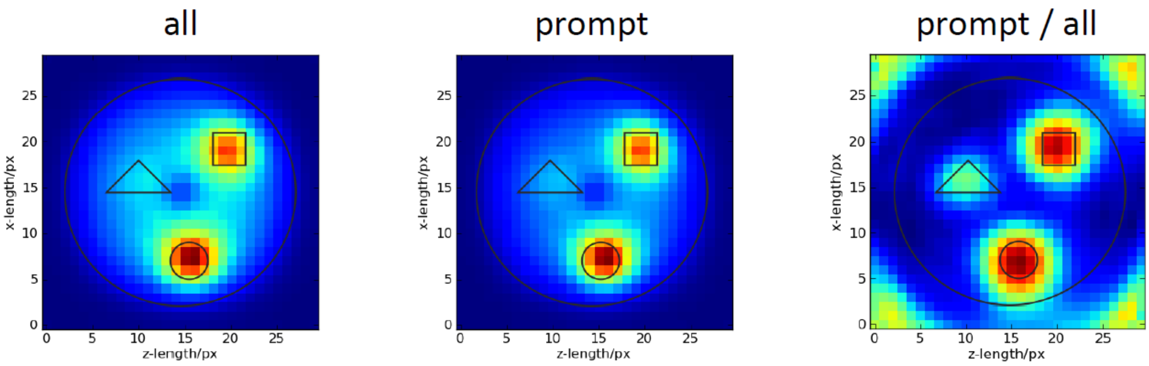

Figure 5. Reconstructed distribution of annihilation events for different regions of annihilation lifetimes after thirty iterations of the MLEM algorithm. Left picture shows all measured annihilations while the central picture is gated on a time interval between –1 ns to 2 ns (prompt). The right picture 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 (MLEM) algorithm [12] has been employed. The algorithm allows reconstructing iteratively a 3-D distribution of events from the set of all measured annihilation events and the system matrix which comprises all possible combinations of detector pixels (169 × 169), rotation angles (180), image voxels (30 × 30 × 30) resulting in an overall size of 138 billion matrix cells. Two reconstructed distributions which are obtained from full (Fig. 5, left) and gated on only prompt annihilation events (Fig. 5, centre) are again used to generate a contrast enhanced annihilation-lifetime sensitive source distribution shown in Fig. 5, right side.

The newly developed system complements earlier developments of a positron annihilation microprobe which enabled high-resolution two-dimensional defect analysis at surfaces [13, 14] or a high-energy positron beam for PALS studies in bulk material [15]. Further developments of the presented system aim at improving the position resolution by using smaller scintillator crystals and improved time resolution by employing digital silicon photomultiplier from Philips Digital Photon Counting which showed an improved time resolution already [16].

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