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Positron annihilation lifetime spectroscopy at a superconducting electron accelerator

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Abstract. The Helmholtz-Zentrum Dresden-Rossendorf operates a superconducting linear accelerator for electrons with energies up to 35 MeV and average beam currents up to 1.6 mA. The electron beam is employed for production of several secondary beams including X-rays from bremsstrahlung production, neutrons, and positrons. The secondary positron beam after moderation feeds the Monoenergetic Positron Source (MePS) where positron annihilation lifetime (PALS) and positron annihilation Doppler-broadening experiments in materials science are performed in parallel. The adjustable repetition rate of the continuous-wave electron beams allows matching of the pulse separation to the positron lifetime in the sample under study. The energy of the positron beam can be set between 0.5 keV and 20 keV to perform depth resolved defect spectroscopy and porosity studies especially for thin films.

1. Introduction

Positron annihilation lifetime spectroscopy (PALS) serves as a unique tool for the characterization of lattice defects in materials science. While the annihilation lifetime yields characteristic information about the size of open-volume defects ranging from single atomic vacancies up to even porous microstructures, the kinematical Doppler-broadening of annihilation radiation tells about the local electron momentum distribution at the annihilation site. Long annihilation lifetimes in the order of ns result from production of positronium, especially in porous media and in polymers. The well-established technique of employing radionuclides simultaneously emitting gamma-rays and positrons of typically up to several hundreds of keV is widely applied. Annihilation lifetimes are monotonously related to the void size with a surprisingly low dependence on the material surrounding the void [1]. The quantitative determination of positron annihilation lifetimes nevertheless is hampered when thin

films or layered structures with sub-µm thicknesses which are of high technological relevance are considered. The isotropic emission and the high energy of common positron emitters cause penetration depths which are on the mm-scale. The installation described here overcomes this limitation by using mono-energetic positron beams with kinetic energies between 0.5 keV and 20 keV thus enabling depth-dependent thin film studies on the nm to µm scale [2].

2. The Mono-energetic Positron Source MePS

The Mono-energetic Positron Source MePS has been set up at the superconducting electron linear accelerator ELBE (Electron LINAC with high Brilliance and low Emittance) [3] at Helmholtz-Zentrum Dresden-Rossendorf in a collaborative effort between Martin-Luther University Halle-Wittenberg and the Helmholtz-Zentrum Dresden-Rossendorf. Figure 1 shows the layout of the system schematically. Some of the details of the installation are described below.

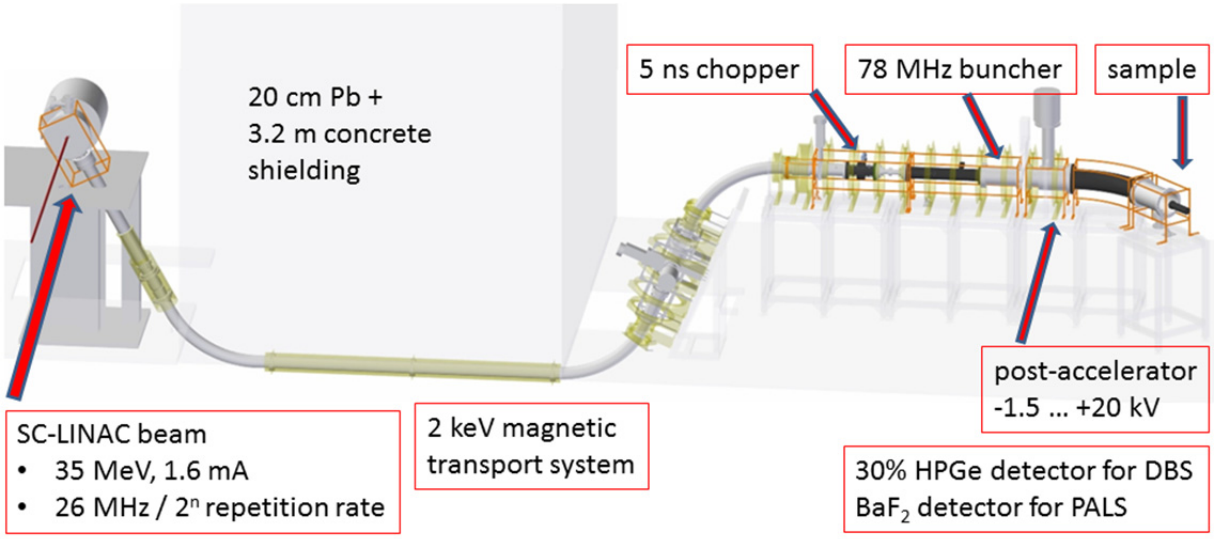

Figure 1. Layout of the monoenergetic positron source.

The incident electron beam is delivered by the superconducting (SC) electron linear accelerator ELBE at energies up to 35 MeV. In contrast to conventional normal conductive accelerators superconducting technology makes possible the application of continuous radio-frequency accelerating electromagnetic fields with electric field gradients up to 20 MV/m. The injection of electrons from an injector can be done in principle with any divisor of the base frequency which is 1.3 GHz for ELBE. However, pulse-preforming by the electron beam bunching system prior to injection restricts the maximum applicable pulse repetition rate to 260 MHz. For positron annihilation studies repetition rates of 1.625 MHz, 6.5 MHz, 13 MHz, and 26 MHz have been employed up to now. The repetition rates have been selected in order to match to the annihilation lifetimes in the samples. Minimization of the effect of pulse-overlap (lower repetition rate favored) has been done while maximizing the detection efficiency (higher repetition rate favored) at given electron beam bunch charges. If we request a pulse-overlapping fraction of not more than one part in a thousand, maximal annihilation lifetimes should be 90 ns, 22 ns, 11 ns, and 6 ns for the *canonical* repetition rates given above. The electron beam is directed towards a water-cooled tungsten bremsstrahlung converter consisting of 50 foils of 100 µm thickness each with a separation of 100 µm each as shown in Figure 2, left side. The converter is enclosed in a stainless steel casing with attached stainless steel cooling pipes as shown in Figure 2, right side. The converter has been designed to cope with average beam powers of up to 40 kW out of which only 4 kW have been used by now. Electron bremsstrahlung with a continuous spectrum up to the electron beam energy is then converted in turn by pair production into

electrons and positrons inside the converter and also inside the following tungsten moderator. Thermalized positrons from the moderator are being accelerated with a bias potential of +2 kV towards a conductive mesh at a few mm distance. Further positron transport is accomplished by a longitudinal magnetic field of 8 mT with variations of less than 10%.

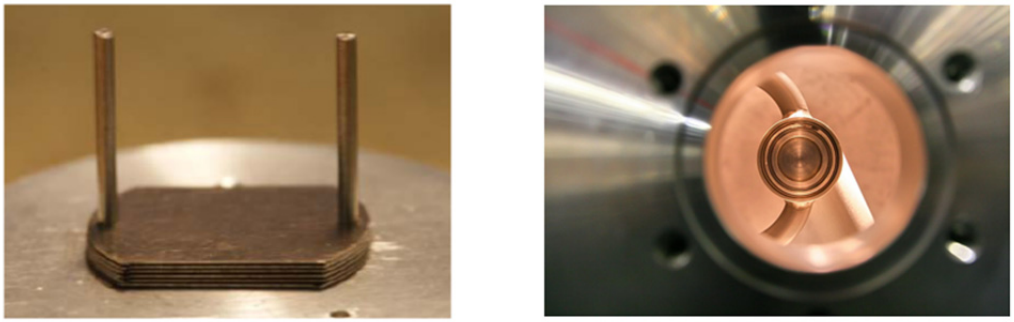

Figure 2. The bremsstrahlung converter. Partially assembled stack of 50 Tungsten foils with 100 µm thickness each (left). Assembled converter as viewed by the electron beam (right).

More than 3 m of iron-enriched concrete and 20 cm of lead shield the accessible laboratory from the positron generating converter. A water-cooled aluminium beam catcher of 600 mm length absorbs most of the radiation penetrating the converter and the moderator. Pure aluminium has been selected in order to minimize photo-neutron production owing the neutron separation energy of 13 MeV for ^{27}Al. The design of the beam catcher has been derived from the liquid lead photo-neutron source which is operated at ELBE, as well [4].

After transporting the beam through the radiation shielding the beam is sent through a double beam chopper which imposes a transversal electric field of up to 500 V_{peak}/cm, see Figure 3 (left). The pulsed electric field has a Gaussian shape with a FWHM of 5 ns while the electrodes are separated by 4 mm. Out-of-phase positrons are deflected towards an adjustable aperture after half a gyration length of 126.3 mm. After removal of those non-phase matched positrons the beam is further longitudinally compressed with a double-slit buncher operating at a frequency of 78 MHz. A resonant circuit drives the central isolated drift tube while generating an electric field strength of up to 2.5 kV/cm at the two gaps, see Figure 3 (right). The two buncher gaps are separated by 509 mm which matches to 3/2 times the buncher period at 2 keV positron transport energy.

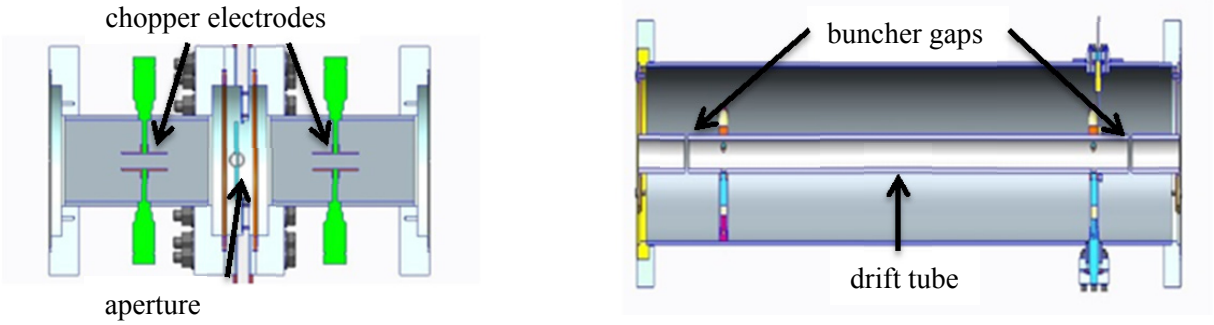

Figure 3. Beam chopper (left) and double-slit buncher (right). The beam enters from the left and additional beamline sections are omitted.

After longitudinal bunch compression by the chopper a 6-stage electrostatic acceleration structure follows which allows the positron kinetic energy and thus the penetration depth inside the sample to be varied. Prior to the target the magnetic guidance system is bent by 45° thus suppressing positrons reflected from the sample and bounced by the accelerator field to impact again onto the sample.

The sample station is kept at a high potential of up to 20 kV. About 3 cm behind the sample a µ-metal shielded BaF_2 scintillation detector is employed for annihilation lifetime measurements. The timing reference is derived from the precision master oscillator of the superconducting accelerator and is phase-matched to the electron bunches of the beam.

A magnetic beam transport system guides positrons to the samples under investigation. A dedicated chopper/buncher system is used to maintain a high timing resolution for depth-dependent annihilation lifetime studies in thin films. The signal-to-noise ratio is above 10^4 while lifetime resolutions of around 230 ps FWHM have been obtained. Figure 4 shows the positron annihilation lifetime of two reference samples with well-known annihilation lifetimes for an incident positron kinetic energy of 10 keV. On the left side, single crystalline Y_2O_3-stabilized ZrO_2 (YSZ) is shown together with a Gaussian distribution with a derived time resolution of 230 ps FWHM. On the right hand side, mono-crystalline silicon is shown. In both cases, the annihilation lifetimes of 181 ps and 218 ps, respectively, which were determined in conventional lifetime studies are well reproduced. Besides the main contribution an additional stable background lifetime of 720 ps of unknown origin is visible which is subtracted in all subsequent analysis. For both samples a signal to noise ratio of $5x10^5$ is obtained.

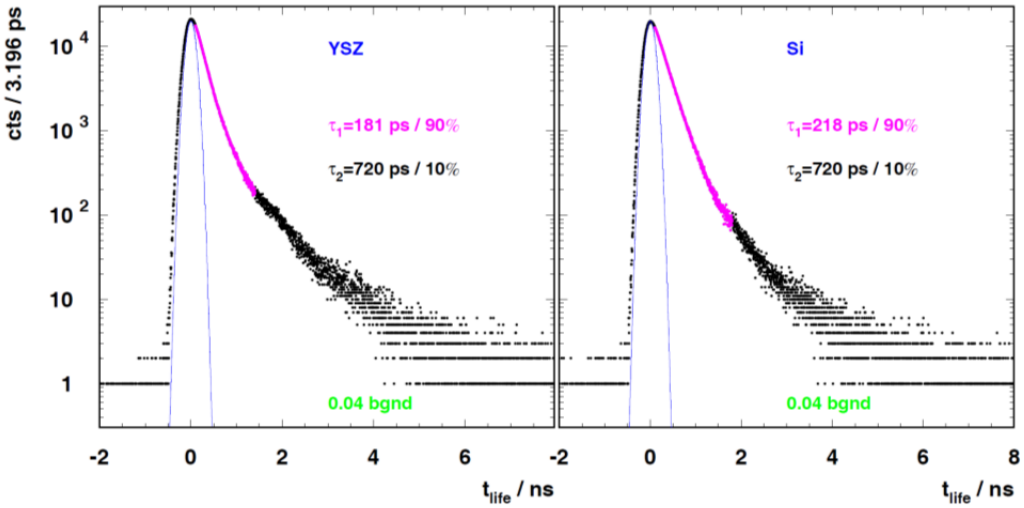

Figure 4. Positron annihilation lifetime distributions for Y_2O_3-stabilized ZrO_2 (left) and wafer-grade silicon (right). A Gaussian distribution with 230 ps FWHM (blue) is included in order to visualize the timing resolution.

An example for longer positron annihilation lifetimes is shown in Figure 5. It shows the variation of ortho-positronium lifetimes with the incident positron energies for nano-porous glasses. The µm-thin glass films have been produced by stimulated phase separation in sodium borosilicate glass into silica and an alkali borate phase thus generating sponge-like porous structures with unknown porosity [5]. Nano-porous glasses feature a tunable pore width and adjustable surface properties which makes them ideal candidates for separation membranes, chemo-sensors, drug delivery, optical coatings, and many other applications. Determination of porosity is hampered when employing conventional porosimetry methods. Both, Hg intrusion or LN_2 sorption methods fail in the case of thin films due to weak signals from small surface areas and the small pore volume. Furthermore, systems with closed porosity cannot be investigated by means of intrusion techniques. In contrast, PALS serves as an ideal tool in such cases. Experimentally, the lifetime distributions are almost free from distortions and the peak to background ratios are between $5x10^4$ and $8x10^4$ due to a slight pulse-to-pulse overlap with the selected beam repetition rate of 1.625 MHz which corresponds to 615 ns pulse-to-pulse spacing. Due to imperfect bunching the timing resolution decreases for positron beam energies below 2 keV. The sample had been covered with a thin carbon layer to prevent positronium from escaping the surface.

Figure 5 shows a scanning electron microscope picture of a sample cut at the left. The nano-porous film has a thickness of about 4 µm.

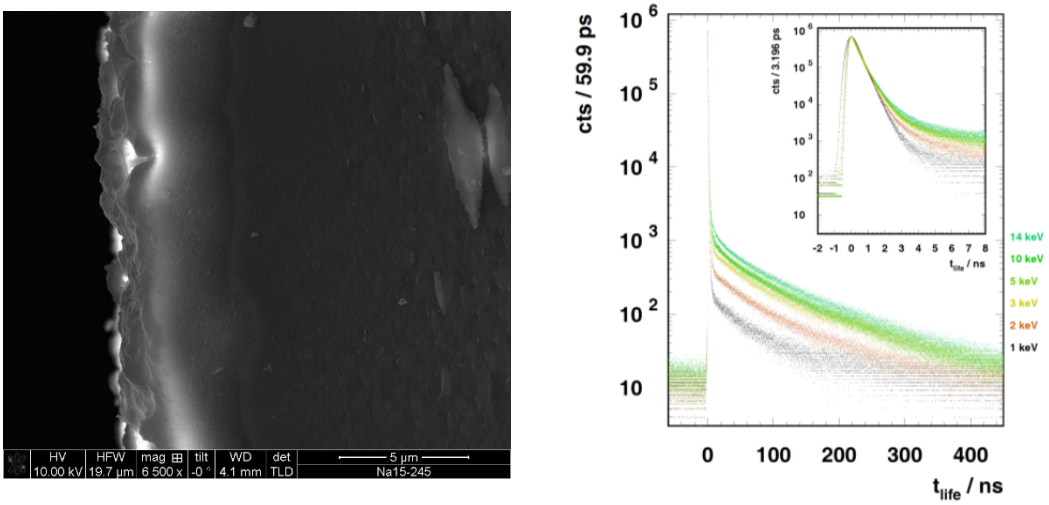

Figure 5. Left panel: SEM picture of the nano-porous film with the surface on the left side and the non-porous structure extending from the centre to the right side. Right panel: Positron annihilation lifetime distributions for nano-porous glasses (inset shown with increased time dispersion).

MePS has also been employed successfully for porosity studies of ultra low-k dielectric thin films as promising materials for advanced inter-connects scheme of ultra-large scale integrated devices. The aim is the reduction of resistance-capacitance delay and cross-talk noise by the introduction of nano-scale voids inside the insulating SiO_2 [6,7]. In the future, MePS will be complemented by an ultra-high vacuum system for in-situ defect studies called Apparatus for In-Situ Defect Analysis (AIDA). Here, the introduction and the annealing of defects during ion-irradiation, temperature treatment and during thin film growth will be studied.

3. The Gamma-induced Positron Source GiPS

A second accelerator-driven positron source is realized in the so-called Gamma-induced Positron Source (GiPS) [8]. A pioneering technology called "accelerator-based γ-ray-induced PAS" or AGAPS at a normal conducting electron LINAC had been developed at Idaho State University and it was used for Doppler-broadening spectroscopy experiments [9]. The setup employs high-energy bremsstrahlung for pair production right inside the sample under study. The GiPS setup additionally allows annihilation lifetime experiments, as well, and it is especially suited for extended bulk samples, or samples which cannot be exposed to external positrons sources like positron beams or radioactive sources or because they are imposing hazardous conditions (high pressure, high temperature, intrinsic radioactivity), or if the sample handling imposes difficulties (fluids, gases, organic samples). Derived from a setup for nuclear resonance fluorescence studies [10] special emphasis has been put on background reduction and shielding. Recently, positron annihilation lifetime studies on point defects in the skyrmion-lattice compound MnSi [11] and on the origin of luminescence and scintillation in ZnO [12] have been published. As for the MePS source the beam repetition rate can be matched to the positron annihilation lifetime thus optimizing for high average intensity and low detector pile-up distortions.

4. The Slow Positron System of Rossendorf

While the accelerator-driven positron sources require sharing beam time with users of beam lines, complementary Doppler-broadening spectroscopy studies are being performed at the ^{22}Na-based

mono-energetic positron beam SPONSOR (Slow POsitroN System Of Rossendorf) [13]. Here, Doppler-broadening studies are performed with higher statistics and more energy steps. Additionally, with both Germanium detectors arranged face-to-face and perpendicular to the beam line coincidence Doppler-broadening experiments are performed which allow for a two order of magnitude improvement in signal to noise ratio. ^{22}Na-based lifetime experiments for bulk materials can be performed at a conventional lifetime setup. As a precursor for the MePS installation a pre-stage of AIDA has been implemented at the SPONSOR beam and in-situ experiments on Fe-Al alloys have been performed [14].

5. Conclusion

The mono-energetic positron source MePS and the Gamm-induced Positron Source GiPS represent the first accelerator-based setups for positron annihilation lifetime, Doppler-broadening and age-momentum correlation measurements operated in a continuous wave mode. Adjustable beam repetition rates are selected to match annihilation lifetimes in order to obtain efficient measurements and low pile-up distortions. Conventional positron sources complement the accelerator-based setups for in-depth and precursor studies.

Acknowledgements

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