

# Operation and testing of the PULSTAR reactor intense slow positron beam and PALS spectrometers

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**Abstract.** An intense slow positron beam has been established at the 1-MW PULSTAR nuclear reactor. The beam is operational generating mono-energetic positrons with an energy of 1-keV. The maximum measured intensity slightly exceeds  $10^9$  e+/s. The beam is operated routinely with an intensity of approximately  $5 \times 10^8$  e+/s. The positrons are generated through gamma-ray pair production interactions in two back-to-back banks of tungsten converter/moderators. The gamma-rays are produced in the PULSTAR core and by thermal neutron capture in a cadmium shroud that surrounds the tungsten. The primary utilization of the PULSTAR positron beam is the characterization of nanoscale structure in materials. Consequently, the beam has been equipped with two state-of-the-art PALS spectrometers. The first spectrometer is dedicated to measurements in materials such as metals and semiconductors. This spectrometer is based on pulsing and bunching of the primary beam and is currently operating with a timing resolution of approximately 390 picoseconds. The second spectrometer is dedicated to measurements in materials where positronium formation is promoted. The timing resolution of this spectrometer is designed to be  $\sim 0.5$  nanosecond with an on-sample spot size of 1-2 mm. For both spectrometers, the energy of the positrons can be varied to allow depth profiling with on-sample intensity exceeding  $10^6$  e+/s.

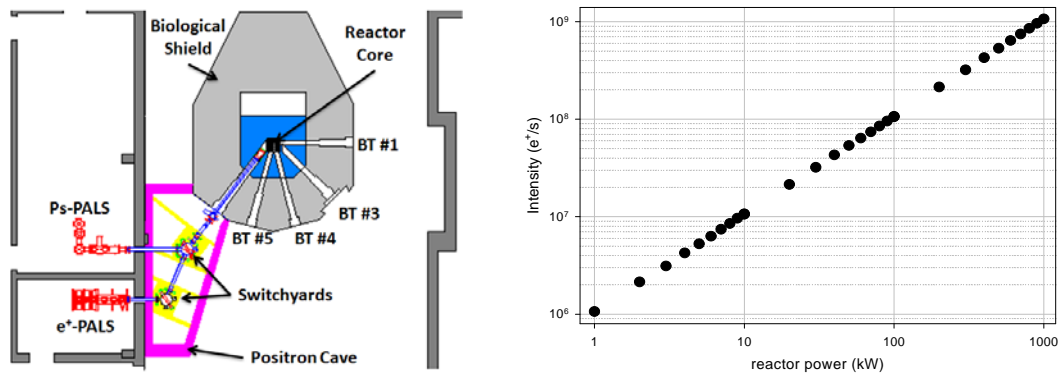
## 1. Introduction

The use of positrons and positronium has proven to be a powerful nondestructive technique to characterize the properties of materials at the nanometer scale. The utilization of low energy beams of positrons wherein the positron's implantation depth can be controlled vastly expands the possible applications by enabling the study of thin films, surfaces, interfaces, and depth-dependent phenomena [1,2]. In particular, beam-based positron spectroscopies have demonstrated great utility in characterizing nanoporous thin film materials and nanocomposites [3]. Introducing and controlling nanometer-sized pores is a rapidly developing area in nanotechnology with wide-ranging implications from H storage and CO<sub>2</sub> catalysis to next-generation interlayer dielectrics in microchips [4]. Depth-profiling will be essential to understanding the role of free volume in: (1) controlling transport properties in polymer electrolytic membranes in fuel cells and thin polyamide control layers in reverse osmosis membranes; (2) correlating with electrical properties in organic semiconductors; and (3) determining thermal and mechanical properties of thin polymer films and nanocomposites. Despite this promising and demonstrated potential, low positron beam rates and/or poor accessibility to beams have limited widespread adoption of positron techniques. To address such issues a new high rate

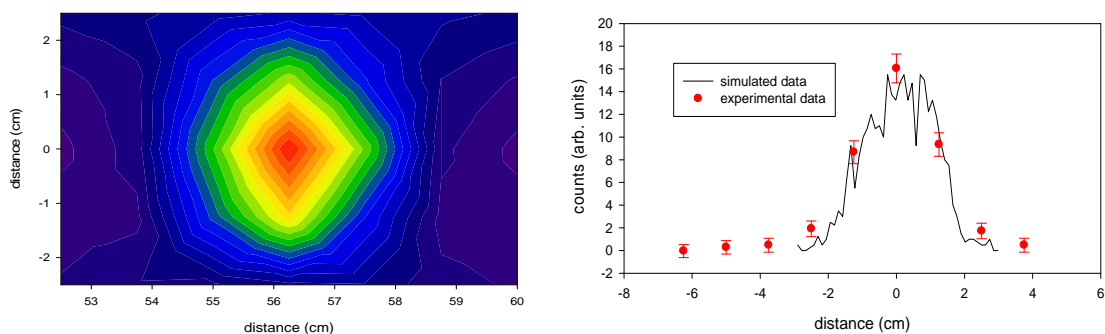
positron beam attached to two different positron/positronium lifetime spectrometers has been constructed at the PULSTAR nuclear reactor on the campus of North Carolina State University.

## 2. The Intense Slow Positron Beam

The positron beam-line is located in beam tube number 6 of the PULSTAR reactor. This beam is currently operational, generating quasi mono-energetic positrons with a mean energy of around 1 keV and a spread of nearly 30 eV. To date the maximum measured intensity slightly exceeds  $10^9$  e<sup>+</sup>/s and the beam is operated routinely with an intensity of approximately  $5 \times 10^8$  e<sup>+</sup>/s. The positrons are generated through gamma-ray pair production interactions in two back-to-back banks of tungsten converter/moderators. Each tungsten bank is made of interlocking tungsten strips (with 1-cm pitch) and is 22-cm in diameter and 2.5 cm thick. Each individual bank underwent four hours of vacuum annealing at 2200 K. The moderators are placed within an aluminum vacuum chamber that is located at the end of a magnetic solenoid tube and inserted into the 30-cm x 30-cm square beam tube. Gamma-rays produced by the core interact in the moderators producing positrons via pair production reactions. In addition, 0.25 mm thick sheets of cadmium, which shroud the tungsten moderator portion of the vacuum chamber, are used to harden and intensify the gamma-ray spectrum by generating gamma-rays through thermal neutron (n,γ) capture reactions. Positrons extracted from the moderators are focused and accelerated by a six element electrostatic lens system to an energy of 1 keV. Subsequently, the positrons are injected into a 60 Gauss (0.006 T) magnetic solenoid for transport outside the reactor biological shield and through two magnetic beam switches allowing the beam to be directed into either of two positron annihilation lifetime spectrometers (PALS) spectrometers. Figure 1 shows the intense positron beam facility (including the spectrometers) and the measured intensity of the generated positrons. Figure 2 shows the image and profile of the beam exiting the beam-line and into the spectrometers. The FWHM measured in a field of 80 G is 2.3 cm.



**Figure 1.** The layout of the slow positron beam facility including the two PALS systems (left). The positron count rate as a function of reactor power (right).

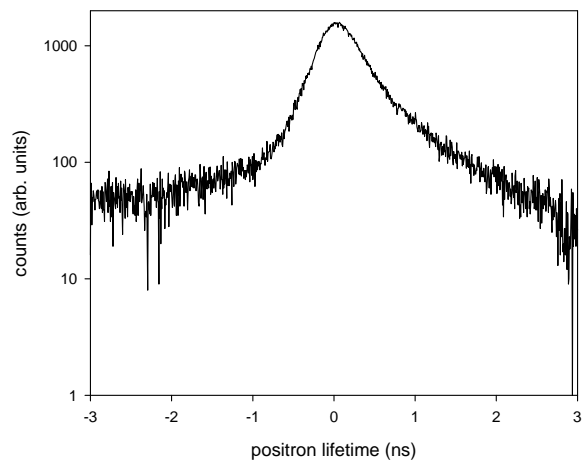
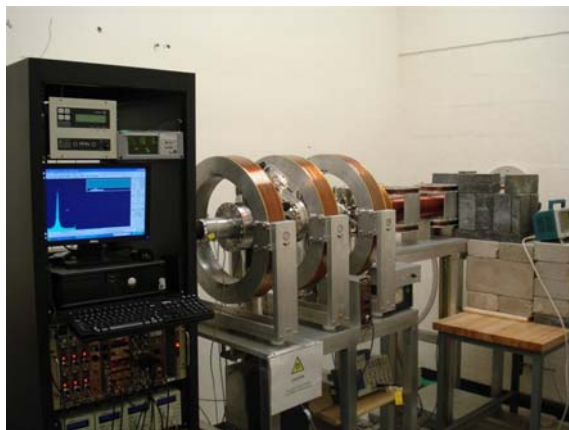


**Figure 2.** An image of the beam at the end of the beam-line (left). The beam profile of the beam as compared to simulations (right).

### 3. The lifetime spectrometers

The first of the two spectrometers is dedicated to measuring positron lifetimes on the order observed in materials such as metals and semiconductors (i.e., lifetimes in the range of 300 picoseconds or less). This instrument utilizes time varying electric fields to generate narrow bunches (< 300ps) of positrons with a 50 MHz rep. rate and the beam is magnetically guided to the sample, which can be biased to negative potentials to vary the implantation energy. Positrons entering the spectrometer are first implanted into a 1 cm diameter, 100 nm thick single crystal tungsten remoderator foil to which a 50 MHz ramp voltage is applied. This acts as a pre-buncher for the main bunching stage. The velocity modulation is optimized by changing a DC offset placed on the moderator ensuring that the focal point of the pre-buncher is at the entrance of the main buncher. This is a double gap device comprising a 9 cm long tube with a 50 MHz, 280 V sine wave, applied to achieve time focussing at the sample. The length of the bunching electrode was calculated so that positrons entering the buncher exit in phase with the applied sine wave, the amplitude of which is chosen to result in optimum time focusing at the sample.

At present, the main buncher has been tested with the remoderation stage withdrawn, i.e., using positrons from the primary beam. Since these positrons have a wide range of transverse energies a retarding potential was applied to a repeller tube prior to the buncher to reduce the longitudinal energy spread at the buncher and improve the time resolution. A lifetime spectrum for Si has been collected using this system and is shown in Figure 3 along with a picture of the spectrometer as it currently stands. The spectrum was fitted with the PALSFIT [5] software giving a resolution estimate of 390 ps.

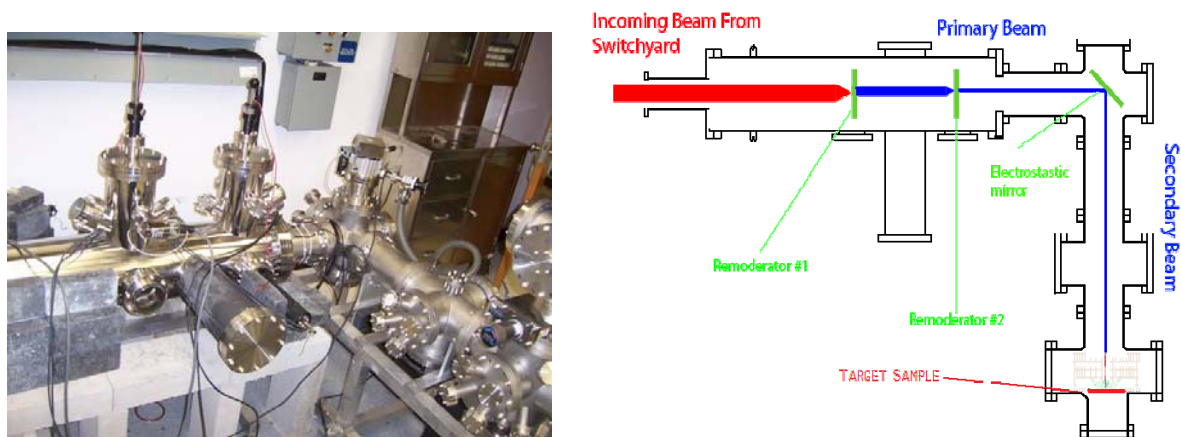


**Figure 3.** The positron lifetime spectrometer (left) and a lifetime spectrum obtained in Si (right).

The second spectrometer is dedicated to the measurement of lifetimes in materials where positronium formation is promoted. In this case, the main positron beam is accelerated to 8 keV after it leaves the magnetic field and is electrostatically focused to a diameter of 20 mm on a 100 nm thick tungsten transmission foil remoderator (the small reduction in beam diameter from 30 mm to 20 mm is due to the large (500 eV) spread in the longitudinal beam energy in the 0.006 T guiding field). After acceleration to 5 keV the remoderated beam is focused to a 1-2 mm spot on a second transmission foil remoderator and after further focusing the final beam on target can be ~1 mm diameter at even the lowest beam implantation energies. The predicted on-target positron rate is  $5 \times 10^6$  /s, which is still sufficient for the spectrometer to be dead-time limited in the individual pulse-counting mode. Random background rates further limit the overall coincidence data rate to  $\sim 10^5$  events/s. A typical lifetime spectrum will thus be acquired in less than one minute.

The start signal for the lifetime measurement using this spectrometer is obtained from a channel electron multiplier array (CEMA) detecting secondary electrons, emitted when positrons impact the target. The positron beam passes through a co-axial hole in the CEMA and strikes the sample target

about 1 cm behind it. A subsequent annihilation gamma-ray is detected in a bank of ten fast-plastic scintillation detectors and a time digitizer measures and records the time interval for each event. Timing resolution is limited to approximately 0.5 ns due to the energy and angular distribution of the reemitted secondary electrons but this is acceptable for measurement of positronium lifetimes in polymers and insulators. Figure 4 below shows this spectrometers current setup.



**Figure 4.** The Ps-PALS spectrometer (left) and a schematic showing the main components (right).

#### 4. Conclusions

An intense slow positron beam has been established at the PULSTAR nuclear reactor of North Carolina State University. This beam operates with a routine intensity of around  $5 \times 10^8$  e<sup>+</sup>/s, a diameter of 2.3 cm FWHM and an energy of 1-keV with an overall spread of approximately 30 eV. Two positron/positronium lifetime spectrometers are currently undergoing testing. The first spectrometer has demonstrated a time resolution of 390 ps and further improvements are anticipated. Doppler broadening techniques can also be performed with each spectrometer. Once completed, this facility will provide a positron beam with energies up to 30 keV and a millimeter spot size for applications in nanophase characterization of materials.

#### References

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