

DEVELOPMENT OF PHOTON-INDUCED POSITRON ANNIHILATION LIFETIME SPECTROSCOPY USING AN S-BAND COMPACT ELECTRON LINAC*

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Abstract

A new photon-induced positron annihilation lifetime spectroscopy approach has been developed using an S-band compact electron linac at the National Institute of Advanced Industrial Science and Technology (AIST). The high energy (<42 MeV), intense (10^9 photons pulse⁻¹), and ultrashort pulse (3 ps pulse width) photon beam creates positrons throughout an entire sample via pair production. A positron lifetime spectrum can be obtained by measuring the time difference between the accelerator's RF frequency and the detection time of the annihilation gamma rays. The positron lifetime for yttria-stabilized zirconia has been successfully measured.

INTRODUCTION

Positron annihilation lifetime spectroscopy (PALS) is a very sensitive tool to characterize materials and study crystal lattice defects like vacancies, dislocations, and clusters at the nanometer scale. The positron lifetime in a material depends on the electron density around the positron. If positrons are trapped in vacancies, the positron lifetime becomes long. Conventional approaches to PALS have been developed using positron sources, such as ²²Na [1], or slow positron beams generated by accelerators [2]. However, PALS has been restricted to thin samples (a few mm) because of the limited range of the positron in materials. In many industrial applications, it is important to know such defects for the entire sample volume.

PALS for thick samples is possible by using high-energy photons to create positrons inside a sample via pair production. The method of photon-induced positron annihilation lifetime spectroscopy (PiPALS) has been previously demonstrated at several facilities. At Idaho State University, proton capturing high-energy coincidence gamma rays through the ²⁷Al(p, g)²⁸Si reaction were employed [3-4]. At Hermholtz Zentrum, ultrashort bremsstrahlung photon pulses generated by the ELBE superconducting linac were used [5].

We have developed a PiPALS system using an electron linac with a photocathode RF gun system at the National Institute of Advanced Industrial Science and Technology (AIST) [6]. Intense, ultrashort photon pulses with energies up to 42 MeV can be generated. Since the 3 ps

pulse width of the ultrashort photon pulses is negligible compared to typical positron annihilation lifetimes (0.1 to 1 ns range), PiPALS is realizable. In this paper, we report the experimental results of PiPALS using the S-band compact electron linac.

EXPERIMENTAL SETUP

The S-band compact electron linac at AIST consists of an injector, a linac, and an achromatic arc section. The injector consists of a Cs₂Te laser photocathode RF gun with an S-band 1.6 cell cavity and a solenoid magnet for emittance compensation. The linac has two 1.5-m-long S-band accelerating tubes which have a $\pi/2$ mode standing wave structure. A high quality electron beam is emitted via the photoelectric effect by injecting the laser into the photocathode. The charge, bunch length, and accelerated energy of the electron beam generated from the injector are 1 nC bunch⁻¹, 3ps (rms), and 4 MeV, respectively. The time jitter between the S-band RF accelerating frequency (2856 MHz) and the laser is approximately 0.1 ps. The electron beam can be accelerated up to approximately 42 MeV with the S-band accelerating tubes. The electron beam then passes through the achromatic arc section and bends through 90-degrees to reduce the background because of the dark current from the linac. The energy spread of the electron beam is 0.2% (rms). A 3-D bunch shape of the electron beam maintains the pulse shape of the laser, which has a Gaussian distribution.

A schematic of the PiPALS experiment is shown in Fig. 1. Ultrashort photon pulses were generated via bremsstrahlung radiation when the electron beam passed through a 0.5 mm thick tungsten plate. The temporal structure of the ultrashort photon pulses is maintained during the process of bremsstrahlung radiation because the tungsten plate is thin. The spatial acceptance of the ultrashort photon pulses was restricted by a collimator. The collimator was composed of a 150 mm thick lead block with a 2 mm diameter hole. The parameters of the electron beam and the ultrashort photon pulses are summarized in Table 1.

A Monte Carlo simulation for the ultrashort photon pulses was conducted using the EGS5 code [7]. EGS5 is a Monte Carlo simulation code for the transport of electrons and photons. The energy and spatial distribution of the ultrashort photon pulses were calculated via a simulation in EGS5 of the experimental setup. The total number of photons passing through the collimator and having energy

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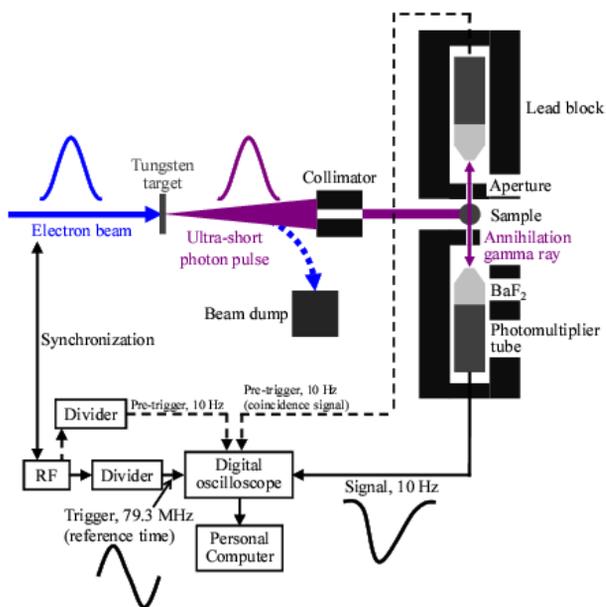


Figure 1: Schematic illustration of PiPALS experiment.

of more than 1.0 MeV was estimated as 1.0×10^5 photons pulse⁻¹ when the electron bunch charge was 1 nC.

Annihilation gamma rays were emitted from a sample and were detected by two barium fluoride (BaF₂) scintillators. To reduce the background, the angular acceptance of the annihilation gamma rays was restricted by an aperture. The aperture was composed of a 50-mm-thick lead block with a 9-mm-diameter hole. The BaF₂ scintillators were covered with Teflon tape and mounted on Hamamatsu photomultiplier tubes (PMTs), H3378 with silicon grease. Each BaF₂ scintillator was 55 mm in diameter and 50 mm thick. The applied voltage of the PMTs was -2400 V.

PALS is realized by measuring the time difference between the annihilation gamma rays and the 36th subharmonic of the RF frequency (79.3 MHz), which corresponds to the mode-lock frequency of the laser. The time of annihilation gamma rays detection was measured by a digital oscilloscope (LeCroy WaveRunner 204 Xi-A)

Table 1: Parameters of Electron Beam and Ultrashort Photon Pulses

Electron beam	Energy	42 MeV
	Bunch charge	1 nC
	Bunch length	3 ps (rms)
	Beam size	50 μm x 50 μm (rms)
	Repetition rate	10 Hz
Ultrashort photon pulse	Energy	< 42 MeV
	Number of photons	1.0×10^5 photons pulse ⁻¹
	Pulse width	3 ps (rms)

which has 4 input channels, a sampling rate of 5 GS/s, and a bandwidth of 2 GHz. A divided signal of 10 Hz from the RF frequency and the annihilation gamma ray detection signal from one of the two PMTs supplied a pre-trigger signal. The RF frequency supplied the trigger signal. The digital oscilloscope converts the waveforms of the coincidence detection signal from another BaF₂ scintillator into text data when the oscilloscope is triggered. The text data were transferred to a personal computer.

Data analysis was conducted on the personal computer. First, waveforms were interpolated via cubic spline interpolation. Second, the energy selections of the interpolated waveforms were carried out. Waveforms corresponding to energies of 0.3 MeV to 0.8 MeV were selected. Third, the selected waveforms were standardized relative to each peak value. Finally, a time distribution was measured when the waveforms cross a 50% slice line of the standardized peak value.

The time distribution represents a positron lifetime spectrum. The lifetime spectrum can be fitted by the convolution integral function $F(t)$ of the lifetime function $I(t)$ and the Gaussian time resolution function $P(t)$. The convolution integral function is expressed as

$$F(t) = \int_0^\infty I(k)P(t-k)dk \tag{1}$$

where

$$I(t) = I_0 \exp\left(-\frac{t}{\tau}\right) \quad \text{for } t > 0$$

$$= 0 \quad \text{for } t < 0 \tag{2}$$

$$P(t) = \frac{1}{\sqrt{2\pi}\sigma} \exp\left\{-\frac{(t-T_0)^2}{2\sigma^2}\right\}, \tag{3}$$

and I_0 is a constant, t is the time, τ is the positron lifetime, σ is the standard deviation, and T_0 is the center time. Eq. (1) turns out to be [8]

$$F(t) = \frac{I_0}{2} \exp\left\{-\frac{1}{\tau}(t-T_0 - \frac{\sigma^2}{2\tau})\right\} \left\{1 - \operatorname{erf}\left(\frac{\sigma}{\sqrt{2}\tau} - \frac{t-T_0}{\sqrt{2}\sigma}\right)\right\} \tag{4}$$

where erf is the error function.

We measured the positron lifetime spectrum of 8-mm-thick yttria-stabilized zirconia (YSZ). Because the YSZ has a single positron lifetime component, we used it to evaluate our photon-induced positron annihilation lifetime spectrometer.

RESULTS AND DISCUSSION

Experimental data are shown in Fig. 2. The positron lifetime for the YSZ was found to be $\tau = 162 \pm 18$ ps. The theoretical value of the positron lifetime for YSZ is $\tau = 182$ ps [9]. The experimental value is consistent with the theoretical value. The time resolution of the positron

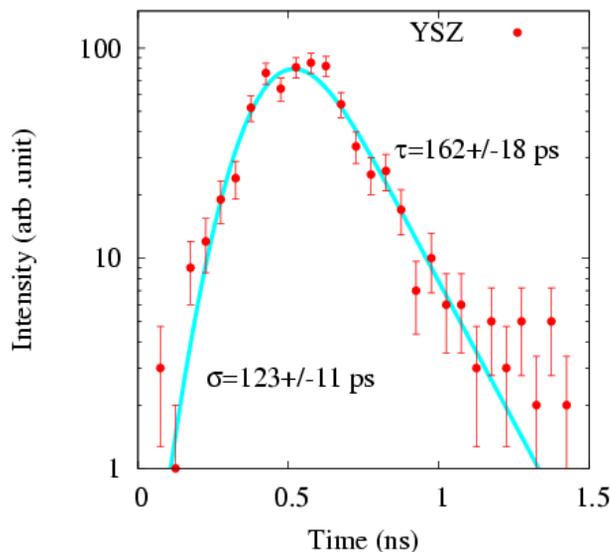


Figure 2: Measured positron lifetime spectrum of yttria-stabilized zirconia (YSZ). Solid curves are least squares fitting curves of data points derived from Eq. (4). Positron lifetime for YSZ is evaluated as 162 ± 18 ps.

lifetime spectrometer was evaluated as $\sigma = 123 \pm 11$ ps (rms) by using least squares fitting. The deterioration of the time resolution has several origins. One of them is the time jitter caused by the variation of the optical path length of the scintillation light due to the geometrical shape of the scintillator. This time jitter can be reduced by using a thin scintillator. However, the detection efficiency of the annihilation gamma rays will be lowered by using a thin BaF_2 scintillator. We plan to use a scintillator with a fast response, high density, and high atomic number (e.g., a Yb^{3+} -doped Lu_2O_3 scintillator).

CONCLUSION

We have developed a photon-induced positron annihilation lifetime spectrometer using the S-band compact electron linac at AIST. The spectrometer consists of BaF_2 scintillators, photomultiplier tubes, a digital oscilloscope, and ultrashort photon pulses based on bremsstrahlung radiation. The time resolution with this setup was evaluated as 123 ps (rms). The positron lifetime for 8 mm thick yttria-stabilized zirconia (YSZ) was measured as 162 ps. The positron lifetimes of bulk materials were successfully measured. We plan to increase the time resolution and measure other samples with short positron lifetimes (e.g., iron). We also plan to increase the repetition rate of the electron beam to 50 Hz to increase the count rate of the positron annihilation lifetime spectrometer.

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