

## Single-shot positron annihilation lifetime spectroscopy with LYSO scintillators



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### ABSTRACT

We have evaluated the application of a lutetium yttrium oxyorthosilicate (LYSO) based detector to single-shot positron annihilation lifetime spectroscopy. We compare this detector directly with a similarly configured PbWO<sub>4</sub> scintillator, which is the usual choice for such measurements. We find that the signal to noise ratio obtained using LYSO is around three times higher than that obtained using PbWO<sub>4</sub> for measurements of Ps excited to longer-lived (Rydberg) levels, or when they are ionized soon after production. This is due to the much higher light output for LYSO (75% and 1% of NaI for LYSO and PbWO<sub>4</sub> respectively). We conclude that LYSO is an ideal scintillator for single-shot measurements of positronium production and excitation performed using a low-intensity pulsed positron beam.

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### 1. Introduction

The development of positron trapping and manipulation techniques [1] has made it possible to produce pulsed positrons from a standard DC positron beam [2]. When a pulsed positron beam is used to generate a positronium (Ps) gas, a burst of annihilation gamma radiation is created. The time and energy characteristics of these photons contain information regarding Ps formation and annihilation processes, and observations thereof can be used to detect Ps–Ps scattering [3], Ps-surface effects in solid-state materials [4] or laser-induced atomic transitions [5–7].

Conventional gamma-ray detectors are usually designed to observe single events and cannot process many thousands of photons arriving in a few hundred nanoseconds. Thus, in order to use such detectors with an intense pulsed beam one has to severely limit its acceptance, losing almost all of the available information [6]. Solutions to this problem were first suggested by Mills Jr. and Platzman [8], and were eventually implemented in a technique called single-shot positron annihilation lifetime spectroscopy (SSPALS) [9]. In this approach an appropriate gamma-ray detector is directly connected to a fast oscilloscope, so that the gamma-ray flux following an intense positron pulse [10] is monitored as a function of time. The detectors that have been used for this are scintillators or Cherenkov radiators optically coupled to a photomultiplier tube (PMT) [11].

This technique was originally developed for studies of Ps–Ps interactions [3], which happen on a short time scale and require intense, high-density, positron pulses [10]. The optimal detector for such measurements is one with a decay constant that is short compared to the 142 ns ground-state Ps lifetime [12], and which has a relatively low light output in order to mitigate saturation of the PMT. Although PbF<sub>2</sub> [11] and heated BaF<sub>2</sub> [13] were shown to be viable options partially fulfilling these criteria, lead tungstate (PbWO<sub>4</sub>, often abbreviated to PWO) proved to have the best overall performance of the materials studied. This technique has made it possible to perform a wide variety of measurements involving Ps produced at both high (e.g., [14,15]) and low (e.g., [16,17]) densities.

Recent work has focused on the production and study of Rydberg Ps atoms using low-intensity, low-density, positron pulses [18–20]. Since these atoms are generally long lived [21], the timing constraints on an appropriate SSPALS detector may be relaxed for Rydberg studies. The ~12 ns decay time of PWO is well suited for the study of short-lived effects since it allows one to integrate lifetime spectra almost from the moment of Ps atom creation. With long-lived Rydberg states, however, the annihilation events of interest may occur on completely different time-scales, and the regions of interest in the corresponding lifetime spectra will change accordingly [22]. Therefore, for measurements of this kind it can be advantageous to use a slower scintillator with a higher light output.

We have tested this possibility using cerium doped lutetium yttrium oxyorthosilicate, (Lu<sub>2(1-x)Y<sub>2x</sub>SiO<sub>5</sub>:Ce). LYSO [23] (as well</sub>

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as its predecessor LSO [24]) has a useful combination of properties, namely its short radiation length ( $\sim 1$  cm), high density ( $8 \text{ g cm}^{-3}$ ), high light output ( $\sim 75\%$  of NaI), and relatively fast decay time (40 ns). As a result these materials have found applications in areas such as positron emission tomography (PET) [25] and high energy physics [26]. Because of its high stopping power LYSO is well suited for studies of low-energy photons [27], which is useful not only in PET scanners [28] but also other positron physics applications (e.g., [29–31]). As we shall discuss, these same properties also make LYSO a good material to use for SSPALS, especially when conducting experiments with Rydberg Ps atoms.

## 2. Experimental arrangement

SSPALS measurements are performed using a pulsed positron source. In the present case we generate a low-density Ps gas by implanting around  $10^5$  positrons in a 5 ns wide pulse into a mesoporous silica film [22]. These materials emit Ps atoms into vacuum following positron bombardment with an efficiency of around 25% [32]. Ground-state Ps atoms are excited to higher lying levels using a pair of Nd:YAG pumped ns pulsed dye lasers: one broadband ( $\sim 100$  GHz) UV laser ( $\lambda \sim 243$  nm) and one narrow band ( $\sim 5$  GHz) IR laser ( $\lambda \sim 750$  nm). Rydberg atoms are produced by a two-step transition via  $n=2$ , as described elsewhere [6,20].

The detector used was a  $10 \times 10$  array of LYSO crystals, each of which was  $4 \times 4 \times 20$  mm. The individual crystals were wrapped on the long sides with reflective Teflon tape, and the resulting  $40 \times 40 \times 20$  mm assembly was optically coupled to a photomultiplier tube (PMT) via a 150 mm long light guide. The light guide was an acrylic cylinder with a 50 mm diameter wrapped in reflective Teflon tape, and was necessary to minimize the effect on the PMT of the  $\sim 100$  G magnetic field used to transport the positrons. For comparison we also used an  $50 \times 38 \times 25$  mm PWO crystal coupled to a PMT with a similar light guide.

The detector positions relative to the Ps production and excitation region are indicated in Fig. 1 and were arranged so that the solid angles subtended by each detector relative to the Ps production region were approximately equal. All of the data presented here were recorded simultaneously from the two detectors, so that the positron and positronium conditions were identical. The PMTs used in each case were different. The LYSO detector was attached

to an EMI type 9954 KA PMT operated with a supply voltage of  $-0.9$  kV. The PWO was attached to a Hamamatsu H10570 PMT, operated with a supply voltage of  $-1.4$  kV. These supply voltages were chosen to avoid PMT saturation.

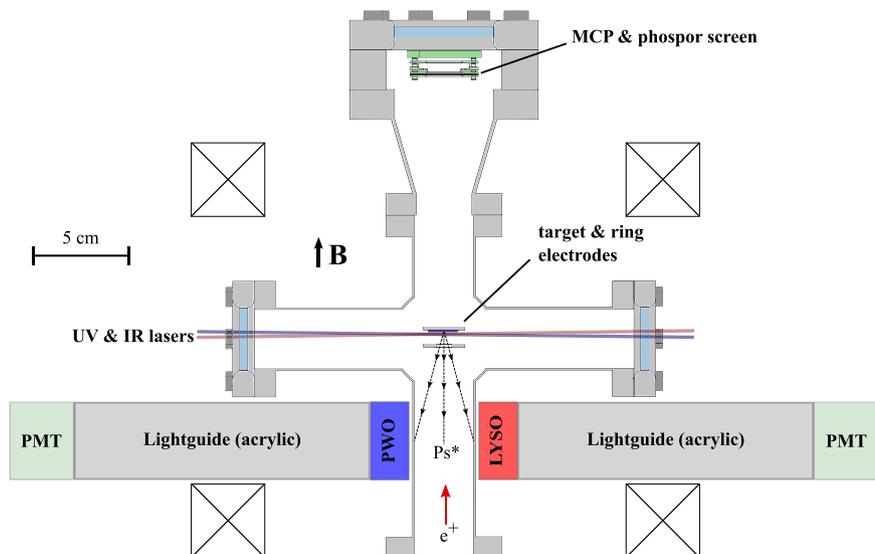
One potential disadvantage of LYSO is that about 2.6% of naturally occurring Lu is the isotope  $^{176}\text{Lu}$ , which is radioactive and has a half-life of  $10^{10}$  years. The decay of this isotope and daughter products lead to the emission of beta particles, and gamma rays with energies of 89, 202, and 307 keV [24]. This can result in an unwanted background signal in PET scanners (for example), but is not important in single shot measurements since the probability of an event occurring in the  $\sim 1$   $\mu\text{s}$  time windows typically used in SSPALS measurements is negligible.

LYSO is available commercially from a variety of vendors, with only slight variations in their properties [33]; the crystals used in this work are of unknown provenance as they were obtained via the secondary market, from an on-line reseller. The crystal dimensions are consistent with those used in PET scanners (e.g., [34]).

## 3. Ground-state positronium

The SSPALS technique involves directly recording the output of a gamma-ray detector with an oscilloscope [9]. The gamma-ray signals obtained in this way are in effect lifetime spectra, examples of which obtained using LYSO and PWO detectors are shown in Fig. 2. These data were recorded with the positron beam implanted into a mesoporous silica film ( $\text{SiO}_2$ ), from which Ps is expected to be emitted, and into a piece of untreated copper (the electrode in front of the silica film), from which we expect almost no Ps to be emitted. The formation of Ps is readily apparent in both detectors as a surplus of counts at later times after the initial peak. This “prompt” peak is due to positrons that annihilate rapidly, either in direct electron–positron interactions following implantation, or via the formation of short-lived (125 ps) singlet Ps atoms [12]. The width of the peak is determined by the width of the incident positron beam and/or the time response of the detector used. Here the positron pulse width is around 5 ns FWHM, and so the observed peak widths are mostly due to the properties of the LYSO and PWO scintillators.

Single-shot lifetime spectra are typically analyzed using the parameter  $f_d$ , defined as



**Fig. 1.** Schematic layout of the target chamber and gamma-ray detectors used in this work. A magnetic field of around 100 G is produced in the Ps excitation region by a pair of coils. An electrode placed in front of the mesoporous silica film allows the electric field in the excitation region to be controlled without affecting the positron beam implantation energy.

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