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Single-shot positron annihilation lifetime spectroscopy using a liquid scintillator

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ABSTRACT

Liquid scintillators provide a fast, single component response. However, they traditionally have a low flashpoint and high vapor pressure. We demonstrate the use of an EJ-309 scintillator (high flashpoint and low vapor pressure variant) to acquire single-shot positron annihilation lifetime spectroscopy spectra using a trap-based positron beam.

INTRODUCTION

Positronium (Ps) is a hydrogenic atom consisting of an antimatter–matter bound state of electronic leptons. The Ps atom is unstable to annihilation with the lifetime dependent on the spin state of the atom (e.g., the ground state: ^1S or p-Ps, 125 ps; ^3S or o-Ps, 142 ns). The photons produced in the annihilation process encode energetic (Doppler broadening) and spin dependent (polarization) information about the electronic environment of the positron. In studies of materials, a variety of techniques are used to obtain atomic-level information via these photons.

Perhaps the most well-known positron annihilation spectroscopy (PAS) technique is positron annihilation lifetime spectroscopy (PALS). Typically, a radioisotope source, such as ^{22}Na, is used to provide energetic positrons (~250 keV), which samples the bulk of a condensed phase material at depths of tens of micrometers. More recently, beam-based PALS techniques have been developed,[1,2] allowing the depth at which positrons are implanted to be varied by varying the kinetic energy of the beam.

Single-shot lifetime spectroscopy, or Single-Shot Positron Annihilation Lifetime Spectroscopy (SSPALS), is a beam-based technique in which an ensemble or a pulse of positrons is injected into a material to produce PALS spectra in one shot, or pulse, of the positron beam. SSPALS was enabled by the development of Surko-style trap-based pulsed positron beams,[3] the subsequent development of intense beams, which included the use of an accumulator,[4]

and the development of fast scintillators.[5,6] Intense positron beam technology allowed the production of dense clouds of Ps atoms in vacuum[7] and the subsequent work in positronium laser physics.[8,9] While positron accumulators and fast scintillating detectors were used in early investigations, SSPALS has been demonstrated without the need for an accumulator or a fast scintillator when investigating vacuum o-Ps.[10]

A key aspect of the optimization of a trap-based positron beam is tailoring the temporal character of the beam pulse. Techniques have been developed to bunch continuous beams,[11,12] but a trap-based beam is typically easier to implement, not requiring tuned RF circuits (e.g., RF buncher). The ejection of positrons from a buffer-gas trap (BGT) can be adjusted to minimize the temporal width of the positron ensemble at a target.[2,13] A typical Surko-style buffer-gas trap can generate positron pulses on a target on the order of tens of nanoseconds without the need for external bunching,[13] as used in this study.

The SSPALS technique can be applied to a range of materials studies in which the lifetime spectrum provides useful information about atomic-level materials properties: mono- and di-vacancies, dislocations, voids, etc. However, this requires both the temporal width of the positron beam and the response of the scintillator to be sufficiently fast. Typically, decay times are longer than the characteristic lifetimes associated with these processes requiring other techniques.[2] SSPALS is particularly useful in materials where the o-Ps (^3S) state is converted to the p-Ps (^1S) state by electron

exchange, resulting in positron lifetimes on the order of tens of nanoseconds. In the case of surface studies, the measurement of the vacuum o-Ps lifetime (142 ns) indicates the emission of Ps from the materials surface. An energy-tunable trap-based positron beam coupled with a fast, single-component detector can provide atomic-level defect information throughout the near-surface (tens of micrometers) volume. Lifetimes of positrons associated with small defects (few nanometers) are less than 1 ns, requiring fast single component scintillators and fast photon detectors for unambiguous identification of defect type.

Liquid scintillators are typically characterized by high flammability and fast scintillation response time with the ability to be used for fast pulse shape discrimination and can have minimum long lifetime afterglow. EJ-309 was developed as a low-flammable alternative to EJ-301. Its pulse shape characteristics provide the ability to distinguish neutrons and gamma rays even in high gamma-ray environments.[14,15] More recently, EJ-309 has been used for traditional or bulk PALS measurements.[16,17] We demonstrate the use of the EJ-309 scintillator for SSPALS and comment on its use in mixed radiation environments.

EXPERIMENTAL METHOD

The positron beam line at the Air Force Institute of Technology (AFIT) was obtained from First Point Scientific, Inc. It consists of a rare gas moderator (RGM) source stage and a Surko-style buffer-gas trap (BGT). A sealed sodium-22 (^{22}Na) source with an activity of 4 mCi was used to provide energetic positrons. The RGM used neon to grow a moderator (neon ice) at 8.2 K. Moderated positrons were magnetically guided from the RGM source (125 G) to the BGT (650 G) where they were trapped, compressed, and cooled to room temperature.[18] Trapping occurs in the first stage of the trap via inelastic (electronic excitation) collisions with molecular nitrogen (N_2). Cooling occurs via inelastic collisions (ro-vibrational excitation) with sulfur hexafluoride (SF_6). The rotating-wall technique was used to compress the positron cloud radially.[19] Positrons were ejected from the trap by dropping the last, or gate, electrode of the trap. The pulse of positrons ejected from the trap was magnetically guided to a gate valve (SS316) used as a beam stop. The annihilation radiation was detected using a 3 in. diameter EJ-309 (510-30 × 30-5) and a 1 in. diameter EJ-309 (510-10 × 10-5) scintillator affixed to a 3 in. photomultiplier tube (PMT) (ET-9821B) operating at −1.5 kV and a 1 in. PMT operating at −1 kV, respectively. The output of these detectors was digitized using a Tektronix DPO7104 Digital Phosphor Oscilloscope (4 channel, 1 GHz bandwidth, 5 GS/s) with the trigger signal provided by the trap control electronics. SSPALS spectra were accumulated on the oscilloscope before being transferred to a computer for analysis.

RESULTS AND DISCUSSION

We acquired SSPALS spectra from our positron beam incident on a 4 $\frac{1}{2}$ in. conflat gate valve constructed from stainless steel (SS) shown in Fig. 1. Both 1 and 3 in. diameter EJ-309 detector assemblies were used to acquire SSPALS spectra simultaneously. These detectors were mounted about the gate valve to maximize the solid angle of the interaction region or approximately the centerline of the positing beam and the center of the conflat gate valve (see Fig. 1).
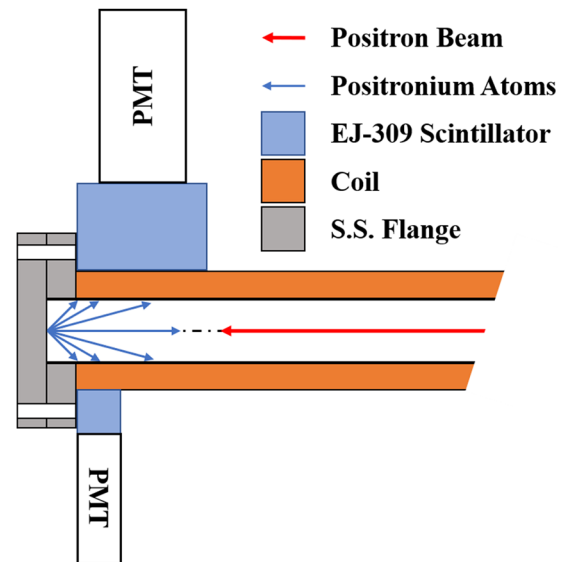

FIG. 1. Diagram of the detector geometry with respect to the positron beam (red arrow), the positronium atoms (blue arrows), and the apparatus.

The results are shown in Figs. 2 and 3 for the 1 and 3 in. detectors, respectively, and are fit using a number of exponentially modified Gaussians (EMG) as discussed below. The results obtained using the 3 in. EJ-309 detector show a shoulder delayed from the peak by ~80 ns. This feature is consistent with the production of Ps atoms at the surface of the gate valve with sufficient kinetic energy to travel to the chamber wall and annihilate.[20] This feature is not obvious in the 1

FIG. 2. SSPALS spectra using a 1 in. EJ-309 liquid scintillator. The black solid line is the measured data, the red solid line is the fit composed of three components, the blue dotted line is the prompt component, the green dashed line is the delayed component, the magenta dotted-dashed line is the o-Ps vacuum lifetime (142 ns), and the black dashed line is a constant background. See the text for discussion of the spectra.
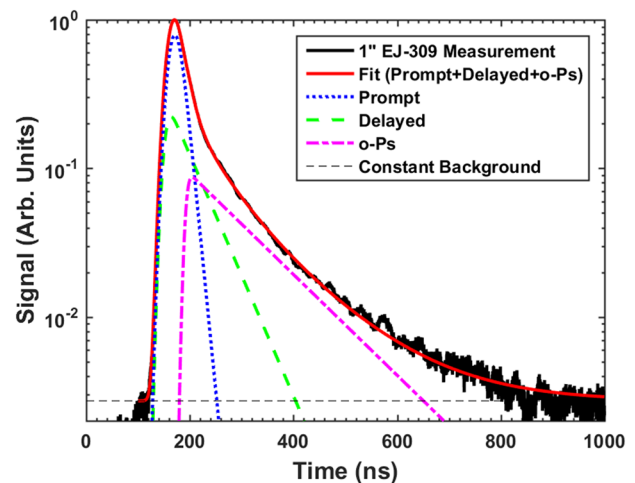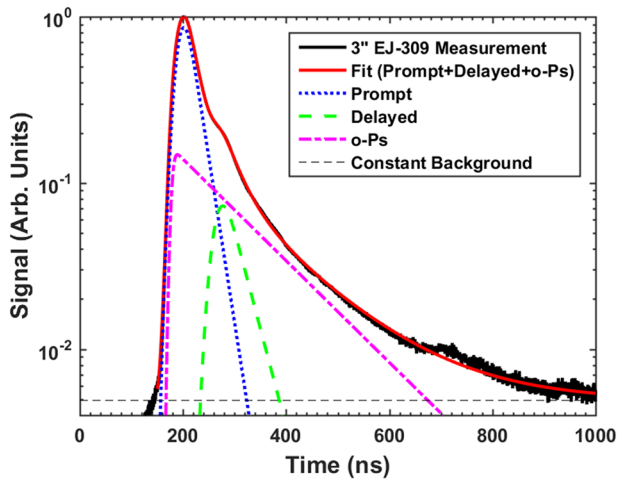
FIG. 3. SSPALS spectra using a 3 in. EJ-309 liquid scintillator. The black solid line is the measured data, the red solid line is the fit composed of three components, the blue dotted line is the prompt component, the green dashed line is the delayed component, the magenta dotted-dashed line is the o-Ps vacuum lifetime (142 ns), and the black dashed line is a constant background. See text for discussion of the spectra.

in. EJ-309 detector due to a greatly reduced solid angle to annihilation events occurring upstream of the gate valve upon the chamber wall.

The spectrum was fit with a function composed of exponentially modified Gaussians (EMG) of the form

$$f(h, \sigma, \tau, \mu; x) = \frac{h\sigma}{\tau}\sqrt{\frac{\pi}{2}} e^{\frac{1}{2}\left(\frac{\sigma}{\tau}\right)^2 - \frac{x-\mu}{\tau}} \text{erfc} \frac{1}{\sqrt{2}}\left(\frac{\sigma}{\tau} - \frac{x-\mu}{\sigma}\right),$$

where h is the height or amplitude, σ is the Gaussian width, τ is the exponential decay constant, and μ is the mean of the distribution. Three of these functions are combined with a constant background

as follows:

$$F = f_P + f_D + f_{o-Ps} + C,$$

where f_P is the prompt component, f_D is the delayed component, f_{o-Ps} is the component due to the decay of f_{o-Ps} in vacuum, and C is the constant background. The prompt peak, f_P, is a convolution of the detector response consisting of the scintillator and the photomultiplier decay time and positron lifetimes on, or below, that order (e.g., para-Ps, pickoff). The delayed component, f_D, is due to any o-Ps that strikes the surface of the chamber and converts to p-Ps.[20] The tail or f_{o-Ps} component is fixed at the o-Ps lifetime or 142 ns. The fit parameters and associated uncertainties are listed in Tables I and II.

The advantage of liquid, and plastic, scintillators is their single component response when considered for use in acquiring a SSPALS measurement. This is in contrast to the response of inorganic crystal scintillators such as BaF$_2$;[21] see Table III for a comparison of various scintillators.[22–36] Suppression of the slow component of the BaF$_2$ fluorescence was investigated[5] and a fast detector based on Cherenkov radiation in a lead fluoride (PbF$_2$) crystal[6] but lead tungstate (PbWO$_4$) is typically used.[7] However, lead tungstate is not a single component scintillator but has many slow components that can be suppressed via doping (see Ref. 30 and references therein). These components are significantly smaller in amplitude than the slow component in BaF$_2$, which allows PbWO$_4$ to be successfully used for SSPALS[1] when only the fraction of vacuum o-Ps is required typically in laser exaction of o-Ps experiments.

The other criterion for choosing PbWO$_4$ as a scintillator for SSPALS is its low light output.[6,7] This is particularly true for systems in which large numbers of annihilation events occur, leading to saturation of the PMT. This is particularly true when a positron accumulator or a strong source ($\geq$50 mCi) is used. When a low-intensity source is used (e.g., <10 mCi) without an accumulator, scintillators with higher light output can be used without saturation of the PMT while maintaining a useful solid angle or higher repetition rates can be used. SSPALS has been demonstrated using an lutetium yttrium oxyorthosilicate (LYSO) scintillator,[10] which has

TABLE I. Fit results for the 1 in. EJ-309 SSPALS spectrum shown in Fig. 1. The o-Ps lifetime, in bold-face, is fixed.

	Prompt	Delayed	o-Ps
h (arb. units)	1.0236 ± 0.0046	0.8386 ± 0.0056	0.7885 ± 0.0100
τ (ns)	12.8379 ± 0.0694	58.2456 ± 0.5554	**142**
σ (ns)	11.3883 ± 0.0333	8.8798 ± 0.0399	5.6224 ± 0.0614
μ (ns)	162.4672 ± 0.0344	150.2082 ± 0.1295	191.6812 ± 0.0782

TABLE II. Fit results for the 3 in. EJ-309 SSPALS spectrum shown in Fig. 2. The o-Ps lifetime, in bold-face, is fixed.

	Prompt	Delayed	o-Ps
h (arb. units)	1.3754 ± 0.0020	0.1354 ± 0.0010	1.7145 ± 0.0072
τ (ns)	21.8213 ± 0.0371	36.9062 ± 0.2779	**142**
σ (ns)	12.1052 ± 0.0106	15.1065 ± 0.0955	5.3893 ± 0.0239
μ (ns)	189.9908 ± 0.0113	260.2338 ± 0.0812	177.2024 ± 0.0356

TABLE III. List of scintillating materials and their timing parameters. Scintillators listed in parenthesis are equivalent materials.

Scintillator	Phase	Decay time (ns)	References
NaI(Tl)	Crystal	250	23
BGO	Crystal	300	25
LYSO	Crystal	45	24
PdWO$_4$	Crystal	1–50, 100, 500	31, 35, and 36
BaF$_2$	Crystal	0.6, 630	26
LaBr3:Ce	Crystal	63	22
BC-408 (EJ-200)	Plastic	2.1	27
BC-412 (EJ-208)	Plastic	3.3	27
BC-418 (EJ-228)	Plastic	1.4	34
BC-420 (EJ-230)	Plastic	1.5	34
BC-422[a] (EJ-232[a])	Plastic	1.6	34
EJ-309	Liquid	3.5	37
EJ-276	Plastic	13, 35, 270	32

[a]Quenched versions of BC-422(BC-422Q), EJ-232 (EJ-232Q), provide faster decay times.[28,33]

a significantly higher light output and longer decay time but only contains a single component.

The EJ-309 liquid scintillator was developed by Eljen Technologies as a replacement for EJ-301, a xylene based liquid scintillator with a low flash point (26 C) and high vapor pressure (6 T at 20 C). It is typically used for fast neutron spectroscopy and is similar to NE-213. The advantage of EJ-309 is primarily safety as it has a higher flash point (144 C) and a significantly reduced vapor pressure (0.002 T at 20 C).[37] The EJ-309's sensitivity to annihilation gamma rays and fast decay time allow it be used as a detector for SSPALS.

Liquid scintillators allow exotic geometries to be more easily fabricated than plastic or crystal scintillators. However, liquid scintillators are not vacuum compatible, and thus, compatible plastic scintillators are typically used.[38] The recent demonstration of 3D printable scintillators[29] is a novel approach allowing an arbitrary geometry to be formed. Both 3D printed and liquid scintillators are useful in circumstances requiring a continuous scintillation material about a legacy device, which cannot be mechanically modified or disassembled and where machining is cumbersome. Liquid scintillators provide additional options in the development of instruments for the acquisition of PALS spectra using a tailored positron beam and BGT-based and RF-bunched positron beams. The ultimate timing resolution of the SSPALS implementation is then found to be limited by the timing jitter of the buncher, scintillator, photodetector (PMT), and digital acquisition system bandwidth.

The other advantage of fast liquid scintillators is their pulse shape characteristics.[14,39] This would allow the operation of beam-based PALS techniques, such as SSPALS, to be performed in a range of environments, e.g., nuclear reactors, to monitor materials degradation *in situ*. The utility of PAS techniques to detect defects in nuclear and irradiated materials has been recently reviewed.[40]

CONCLUSIONS

A liquid scintillator (EJ-309) was used to implement Single-Shot Positron Annihilation Spectroscopy (SSPALS) using a buffer gas trap-based positron beam. This demonstration of the SSPALS technique using a liquid scintillator with lower volatility highlights the possibility of using it in new experimental geometries difficult to implement those traditionally used. The fast decay time and single component nature of the liquid scintillator provide superior performance over LYSO in many applications of the SSPALS technique. The pulse shape characteristics allow its use in reactor environments that have a neutron background.

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding authors upon reasonable request.

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