Single shot positron annihilation lifetime spectroscopy

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Recent developments in positron trapping technology have made possible experimentation with dense interacting positronium gases. Along with these capabilities comes a need for suitable measurement techniques, and accordingly we have developed a method to measure positronium lifetimes from a single intense burst of positrons. Our method is based on recording the anode signal from a photomultiplier with a fast oscilloscope following a short-time positron burst which allows us to measure transitory effects as well as high density positronium interactions. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2203336]

Positron annihilation lifetime spectroscopy (PALS) has been widely used for many years as a probe of defects and impurities in solid-state materials[1] as well as to understand their electronic properties.[2] The basic principle of PALS is that a timing spectrometer is activated by a start signal associated with the "birth" of a positron, and the annihilation radiation of that positron is used as the stop signal from which the positron lifetime is determined.[3] Often a radioactive source is used for such work, the most common being ^{22}Na. This isotope emits a beta spectrum of positrons (540 keV end point energy) with each positron quickly (delay ~10 ps) followed by gamma ray of energy of 1.27 MeV that is often used as the start signal in a PALS arrangement.

The use of a positron beam rather than unmoderated beta particles from a source allows a number of refinements to the straightforward PALS setup.[4] For instance, different start signals may be used, such as the emission of secondary electrons from the target or an electrical pulse that is synchronous with the arrival of a burst of slow positrons at the target. Also, by varying the incident beam energy samples may be depth profiled. That is, spectra may be obtained corresponding to implantation at different positions within a sample, which is useful for studying multilayered materials and discriminating between bulk and surface effects. This is sometimes referred to as variable energy positron annihilation lifetime spectroscopy (VEPALS).[4] In all cases, a lifetime spectrum typically containing $(1-10)\times10^6$ events is collected. Depending on the source strength and the details of a particular arrangement, the time taken to obtain a spectrum with adequate statistics can be anywhere from tens of minutes to days.

We report here the development of a technique to record a lifetime spectrum from a single shot from a positron accumulator. The main advantage of this variation of PALS (which we shall refer to as SSPALS) is that the data collection time is defined only by the lifetime to be studied, which then allows for the possibility of measuring transient phenomena. These could be related to the sample or to the positronium itself. As an example of the latter, this technique has already been used to observe the lifetime quenching due to interactions between pairs of positronium atoms confined at high densities in a porous silica sample.[5]

SSPALS relies on the production of large positron pulses with a temporal width that is short compared with the lifetimes to be measured. We generate such pulses using a modified Surko-type positron accumulator[6] similar to that described in Ref. 7. Positrons in the accumulator were compressed radially with a "rotating wall" electric field[8] producing long lived plasmas (>1000 s) containing up to 100×10^6 positrons in a 500 G axial magnetic field.

The accumulator output was focused in time by rapidly applying a potential gradient across the extent of the accumulator electrodes,[9] leading to positron pulses with a duration of 15–20 ns [full width at half maximum (FWHM)]. However, for large numbers of positrons (20×10^6 or more) we found that this width grew (approximately linearly) with the space charge potential, and so bursts containing around 15×10^6 positrons were used.

The data were recorded using a lead tungstate ($PbWO_4$) scintillator attached to an XP2020 photomultiplier. This scintillator material was chosen for its relatively fast decay time (<15 ns). Lead tungstate also has a low afterglow, which proved to be of critical importance in these experiments.

The gamma ray detector was connected directly to an Agilent 54885A 6 GHz bandwidth oscilloscope with a maximum sampling rate of 20 gigasamples/s. Data acquisition was done automatically, with wave forms downloaded to a personal computer (PC) and analyzed essentially in real time. Because the lifetime spectra have a large dynamic (voltage) range, recording the data with a single oscilloscope channel (and hence with a single gain) would have resulted in unacceptably high digitization noise in the low amplitude region. For this reason the output from the photomultiplier was split using a 50 Ω resistive tee and recorded on two channels, with high and low gain. This allowed us to collect the low amplitude part of the signal with maximum gain and still record the "prompt peak." These two wave forms were then spliced together (off-line) to create a single lifetime spectra.

Figure 1 shows single shot lifetime spectra obtained in this manner using a porous silica sample with an average porosity of around 48%. In all spectra a peak is present at around 400 ns. This is due to residual gas atoms (predominantly hydrogen) on the first dynode that are ionized by the primary electron pulse and then returned to the photocathode, generating a second electron pulse.[10] The position of this after pulse depends on the accelerating voltage between the first dynode and the cathode, but generally cannot be

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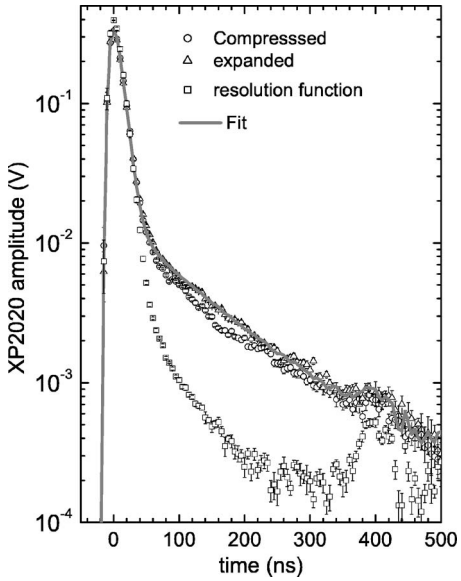

FIG. 1. Single shot lifetimes for the capped TEOS sample and the resolution function. The low density (expanded) data are well fitted using a single lifetime, taking into account the resolution function, but the high density (expanded) data cannot be fitted without additional terms.

changed by more than 50 ns or so without compromising the gain of the photomultiplier. Also shown in Fig. 1 is a spectrum taken with an uncleaned Si sample and a positron energy of 7 keV. This target material will not produce any Ps in the bulk and only a small amount ($<2\%$) of surface Ps is expected at this impact energy. This has been taken as the resolution function of the system and was used in the fitting procedure. This resolution is determined primarily by the width of the positron beam and the decay time of the $PbWO_4$ scintillator; we expect this to be greatly reduced in subsequent work.

The data were taken at low and high densities (expanded and compressed), corresponding to incident positron areal central densities of 0.37 and 3.3×10^{10} cm^{-2}, respectively. The sample was capped with a largely nonporous layer ($\sim5\%$ porosity), and the positron impact energy for these data was 3.5 keV. The low density data were fitted using a single positronium lifetime, while the high density data cannot be fitted without taking into account additional terms related to spin exchange quenching and positronium molecule formation, the details of which are beyond the scope of the present work.[5]

An important difference between conventional PALS and SSPALS is that in the latter the response of the gamma ray detection system is folded into the lifetime spectra. This is in contrast to a leading edge trigger in which only the rise time of the gamma ray detection system is important. Thus, in SSPALS effects such as dynode ringing, cable reflections and rf pickup are invariably present to some degree. The photomultiplier ion after pulse is also an unavoidable feature of such spectra, as shown in Fig. 1. Clearly then it is desirable to minimize such effects, and we have found that a few relatively simple modifications can do so with surprising efficacy. Dynode ringing was reduced by adding 50 Ω resistors in series with the dynode connections in the photomultiplier base; cable reflections were essentially eliminated by careful impedance matching throughout the system. This was particularly important when splitting the signal into two channels as described previously.

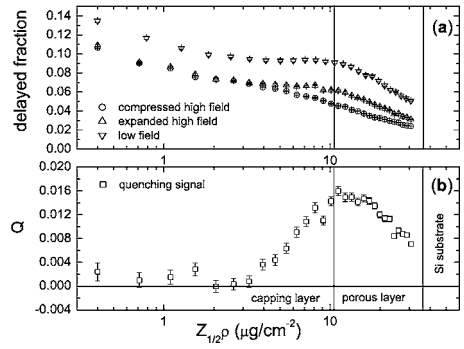

FIG. 2. Variable energy and density single shot positron annihilation lifetime spectroscopy data taken for the same sample used in Fig. 1. In (a) three energy scans are shown, one at high density (3.3×10^{10} cm^{-2}) and one at low density (0.49×10^{-10} cm^{-2}) in the high (1 T) field, and also one for the beam in a low (250 G) field. In (b) the quenching signal Q is shown, which is the difference between the high and low density data of (a).

Another aspect of SSPALS that is not generally important in PALS is detector saturation. With a plastic scintillator we found that it was necessary to use an unacceptably small solid angle to prevent photocathode saturation and a very low photomultiplier gain to prevent anode saturation. Both of these compromises resulted in a large degradation of the signal, with a concurrent worsening of the counting statistics. However, $PbWO_4$ has a naturally low light output [approximately 1% of NaI (TI)] and so the scintillator was placed close to the annihilation region with no saturation problems. Essentially, if enough photoelectrons are present to unambiguously register a single gamma ray, statistical integrity may be retained while minimizing saturation effects. The problem of anode saturation was solved in the usual way by providing larger interdynode voltage drops as one approaches the anode.

The use of a high density pulsed positron beam has also allowed us to develop a different approach to depth profiling in porous Ps forming materials. In conventional VEPALS, lifetime spectra are recorded as a function of the beam energy, and hence implantation depth in the sample. In our methodology this is done at high and low beam densities, and we measure the delayed fraction of a lifetime spectrum in both cases. Because there is an *o*-Ps quenching effect that is directly proportional to the Ps density, we can measure this density as a function of implantation energy (provided there is no significant modification in the Ps density due to diffusion). Figure 2 shows an example of a variable energy and density single shot positron annihilation lifetime spectroscopy measurement made using the same sample as was used to collect the data shown in Fig. 1. The vertical lines in the figure show the nominal thickness of the capping and porous layers as determined by ellipsometry measurements. The data seem to suggest that the capping layer thickness is actually around 7 μg/cm^2 and not 10 μg/cm^2 as shown, which could indicate that the capping layer is more porous than expected. One possible reason for this is that the porosity was determined to be $\sim5\%$ via a gas absorption measurement, which is insensitive to isolated pores. The abscissas of Fig. 2 are determined from the beam energy via the formalism described in Ref. 11, using a density of 2.2 g/cm^3 for nonporous silica.

The delayed fraction in Fig. 2(a) is the integral of the lifetime spectrum from 100 to 300 ns divided by the total, and is a measure of the positronium fraction.[12] It is clear that

for a high density positron beam this fraction is reduced, which is due to spin exchange quenching and positronium molecule formation, both of which lead to *o*-Ps quenching. Even without measuring the rates of these effects directly, the difference between two densities provides a quenching signal that can be used to determine the average Ps density as a function of the average positron implantation depth. This method has the advantage that it is insensitive to vacuum Ps where the chances of Ps interactions are practically zero. Thus, in cases where VEPALS might lead to erroneous data due to long lived vacuum Ps, this method does not. This avoids the need to use some of the usual methods needed to distinguish vacuum from bulk Ps, such as capping the surface or collimating the detector.[13]

We have presented a set of tools that have become available with the introduction of a high density pulsed positron beam system. These methods are certainly not practical alternatives to PALS and VEPALS, and are in fact most useful in circumstances where these techniques cannot be used; they are complementary to them. Furthermore, the nature of SSPALS allows for some unique applications involving transient phenomena. Since the time required to collect a lifetime spectrum is very short (i.e., <1 μs) this could be done in conjunction with an intense laser pulse to observe structural changes in a target on a short time scale. For *temporal* mechanisms such as the formation and annealing of crystalline defects, nucleating of phase transitions, formation and expansion of plasmas upon sudden heating, etc., a series of spectra could be taken to study the time dependence.

The variation of VEPALS we have described has some additional useful properties that have not yet been explored. In particular, the measurement of positronium density in such a direct manner is essentially the single shot analog to probing pore open volumes (and the connectivity to the vacuum) by measuring the relative amount of three-photon annihilations.[13] Also, the relatively small size of the beams used here ($\sim$200 μm FWHM) means that small samples may be used, or crude lateral scanning of larger samples may be performed. With further development we expect the methods described here to become another facet of the toolkit already used for studying the properties of nanoporous thin films and low-k dielectrics.

In addition to characterizing positronium forming materials, we are also able to determine some basic properties of positronium itself, and indeed, this is the main focus of our research. We have already made an observation of Ps–Ps interactions[5] using the approach described here, and with only minor modifications we expect to be able to perform studies of the dynamics of positronium molecule formation on surfaces.

Work is ongoing to improve the timing resolution of the system, which is presently determined by both the positron pulse width and the response of the detection system. Additional bunching can easily create sub-nanosecond positron pulses, and the limiting factor would then be due to the detector used. A Cherenkov radiator (such as PbF_2) can provide essentially instantaneous light, so that when coupled to a single stage microchannel plate photomultiplier we expect to obtain a minimum timing resolution of around 0.5 ns, although this would drastically reduce the count rate.

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[10] We note that the magnitude of the ion after pulse is proportional to the primary pulse to some upper limit, after which it does not increase. We interpret this as being due to complete desorption of the hydrogen from the dynode. This suggests that it may be possible to significantly reduce this feature by using a LED light prepulse just before the main pulse. In other similar work using a much larger signal the ion after pulse in a 56 DUVP PM quickly became negligible. M. Biasini, D. B. Cassidy, S. H. M. Deng, H. K. M. Tanaka, and A. P. Mills, Jr., Nucl. Instrum. Methods Phys. Res. A **553**, 550 (2005).
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[12] This quantity is not the actual positronium fraction because we have not accounted for the magnetic quenching effect; neither have we rigorously defined the integration regions to ensure good differentiation between two and three gamma events. Nevertheless, since the quoted "delayed fraction" is consistently defined for all data it is a useful metric.
[13] See, e. g., D. W. Gidley, K. G. Lynn, M. P. Petkov, J. N. Sun, M. H. Weber, and A. F. Yee, in *New Directions in Antimatter Chemistry and Physics*, edited by C. M. Surko and F. A. Gianturco (Kluwer, Dordrecht, 2001), p. 151.