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A fast detector for single-shot positron annihilation lifetime spectroscopy

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Abstract

When an intense sub-nanosecond positron pulse impinges upon a target, a pulse of γ-rays is created which can yield information concerning electron–positron pairs just prior to annihilation. Many conventional γ-ray detectors are unable to exploit the timing information contained within such pulses, and we describe here the development of a fast detector that is able to do so. Using a single-crystal PbF_2 Cherenkov radiator coupled to a fast photomultiplier tube (PMT), we have produced a detector with a time response of ~4 ns (primarily determined by the PMT response), as well as a low-efficiency detector with a sub-nanosecond response. Since 511 keV photons produce very little Cherenkov light, the problem of photomultiplier saturation is mitigated and this detector is therefore well suited to single-shot positron annihilation lifetime spectroscopy (SSPALS) measurements.

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1. Introduction

Recently, a technique was developed by which positron annihilation lifetime spectroscopy (PALS) [1] may be performed using a single-intense pulse of positrons (SSPALS) [2]. The basis of this type of measurement is that the annihilation γ-rays following a positron pulse are recorded in real time, so that the detector output constitutes a lifetime spectrum. This is fundamentally different from a conventional PALS arrangement in which positron annihilation events are registered one at a time. Thus, in SSPALS, the complete detector response becomes folded into the lifetime spectrum, and consequently the full timing response of the detection system defines the minimum timing resolution of the technique; this inevitably means that the timing resolution of SSPALS is much less than that of PALS, by at least an order of magnitude. Despite its relatively poor timing resolution SSPALS does provide a unique method of studying interactions between positron atoms, which necessarily occur on a time scale of ~100 ns or less. Since DC beams of the required intensity simply do not exist (the pulses we have used have instantaneous currents of ~10 mA) such studies have to be done piecewise, using single-intense pulses, and thus ordinary single-event counting methods cannot be used.

In addition to the detection system response, the time width of the primary positron pulse also affects the achievable time resolution. In Ref. [2], we described a system comprising a positron beam with a width of ~15 ns, and a $PbWO_4$ scintillator which has a decay time of ~15 ns. Together these led to a time resolution of ~20 ns, which is around 100 times worse than a typical PALS system [3]. Experiments in which ortho-positronium (o-Ps) lifetimes that are of the order of the vacuum lifetime (142 ns) are to be measured can be performed with this low time resolution, but there are many experiments where ~nanosecond resolution is required. While it is relatively straightforward to produce a sub-nanosecond beam [4], developing a γ-ray detector with all of the characteristics necessary for SSPALS has proved to be somewhat more difficult and, as is often the case when using scintillators,

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one has to optimize some detector properties at the expense of others. For SSPALS an optimal detector should have a fast time response, a high density, a low light output (to prevent saturation) and minimal afterglow. We report here the results of our efforts to develop such a detector. The data shown were obtained using methods that have been described in detail elsewhere [4]. Briefly, an accumulator is used to form positron plasmas that may contain up to 100 million particles. These plasmas are ejected from an electrostatic and magnetic trap and are bunched into sub-nanosecond pulses. The dump signal is used to trigger an oscilloscope, which then records the anode voltage of a detector that registers the subsequent annihilation gamma rays; this timing information is equivalent to a lifetime spectrum.

2. Detectors commonly used to study positron annihilation radiation

By far, the most common way to detect γ-rays resulting from positron annihilation is via some sort of scintillating material attached to a photomultiplier. There are many different scintillators commercially available [5] with a wide variety of properties. In particular, timing characteristics span a large range, from nanosecond to millisecond, associated with the emission of photons during electronic transitions in the scintillating material.

In order to be useful as a fast γ-ray detector, it is desirable for a scintillator to have a relatively high density and atomic number (and concurrent stopping power) and luminosity, as well as a short decay time. In practice, this means that the radiation length should be less than the size of the crystal (~1 cm or less is ideal) and the light output should be about 20 photons per 511 keV γ-ray. If the light output is too high detector saturation becomes a problem, and if it is too low (i.e., <1) counting statistics will be diminished.

In general denser materials tend to have higher luminosities, but longer decay times. For example, plastic scintillators are often used for fast timing applications, and they can have sub-nanosecond decay times, but the low stopping power means that they are extremely inefficient. Since positrons are experimentally "expensive" this low efficiency is often impractical, and for SSPALS plastic is not a good choice.

A workhorse of positron studies is sodium iodide (NaI) which is a very efficient scintillator [6]. However, with a ~0.2 μs decay time this material is completely useless for SSPALS. When high-energy resolution is required (for example, to study the Doppler broadening of a 511 keV annihilation radiation [7]) solid-state high-purity germanium (HpGe) detectors are usually used. This is not a scintillator; incoming γ-rays create electron–hole pairs, and the charge collection then provides an energy-dependent signal. However, as with NaI, the timing characteristics of these detectors also render them unsuitable for the present purposes.

We used a $PbWO_4$ scintillator [8] for our initial SSPALS experiments [2]. While this was useful for some experimentation, it does not allow us to take full advantage of our sub-nanosecond beam and so a detector with a faster decay time is required. We note that indium-doped single-crystal zinc oxide (ZnO:In) may be a promising sub-nanosecond scintillator [9], but is not yet commercially available. This brief consideration of some commonly used materials is meant only to highlight the fact that all of the desired properties are not usually available in a single material. Derenzo [5] has given a detailed survey of available scintillators.

3. Cherenkov radiation and SSPALS

In contrast to most applications, SSPALS requires that the number of photons entering the PMT cathode per incident γ-ray be low. This is because more than 10,000 events are typically recorded within a few nanoseconds, which can easily saturate a photomultiplier if each event generates many photons. However, in order to preserve the counting statistics there should be, on average, *at least* one photo-electron created per γ-ray. It is not necessary for a low light output to be an intrinsic property of a material attached to a PMT, since filters can be used to limit light transfer to the photocathode.

To obtain the fastest possible timing resolution with a given photomultiplier, one may use a Cherenkov radiator instead of a scintillator since Cherenkov light is emitted instantaneously. Lead fluoride is a good material to use for a Cherenkov detector because it has no detectable scintillation and a high atomic number and density [10]. Cherenkov radiation occurs when a charged particle in a medium moves faster than the speed of light in that medium. That is, $v > c/n$, where v is the speed of the charged particle, c is the speed of light in vacuum and n is the index of refraction of the medium. The most common use of Cherenkov radiation in the detection of γ-rays is based on pair production by high-energy radiation. Particle showers produced by cosmic rays [11] or accelerators [12] generate the signal, but clearly photon energies significantly above 1.02 MeV are required for this. For the observation of low-energy γ-rays below the pair production threshold, the signal is generated via Compton-scattered and photo-ionized electrons in the medium. These electrons stop quickly, and consequently very little light is generated. For this reason, Cherenkov radiators are generally not used for the detection of low-energy γ-rays. However, since the requirement for a high light output is relaxed in this application, PbF_2 is a viable choice.

We may make a crude estimate of the number of photoelectrons generated in a phototube with a PbF_2 Cherenkov radiator attached in the following way. If the incident γ-ray energy is below the threshold for pair production, only bound electrons that are liberated produce Cherenkov radiation. In fact, since the electron

velocity must be larger than c/n the energy threshold for this process in PbF_2 is ~81 keV ($n = 1.76$ at 470 nm).

The number of photons emitted per electron per unit path length, x, between the wavelengths λ_1 and λ_2 is given by [13]

$$\frac{dN}{dx} = 2\pi\alpha\left(\frac{1}{\lambda_1} - \frac{1}{\lambda_2}\right)\left(1 - \frac{1}{\beta^2 n^2(\lambda)}\right). \quad (1)$$

Here N is the number of photons, $\alpha = 1/137$ is the fine structure constant, $\beta = v/c$ and $n(\lambda)$ is the index of refraction of PbF_2 as a function of light wavelength. The number of photoelectrons created per incident γ-ray may be estimated from the sum of the photoelectric and Compton-scattering cross-sections. For 511 keV γ-rays in PbF_2 these are approximately equal ($\sim 7 \times 10^{-2}$ cm^2/g) [14]. However, because of their energy distribution the contribution to the light from Compton scattered electrons is quite low and for this estimate we shall neglect them. This means we will underestimate the number of photoelectrons by, at most, ~20%.

If we evaluate equation (1) using the PbF_2 UV cutoff wavelength $\lambda_1 = 0.25\,\mu$m and a multialkali photocathode 50% cutoff wavelength $\lambda_2 = 0.5\,\mu$m, we find that

$$\frac{dN}{dx} = 917\,\text{cm}^{-1}\left(1 - \frac{1}{\beta^2 n^2(\lambda)}\right). \quad (2)$$

The path length over which Cherenkov radiation can be emitted is set by the ionization energy loss per unit path length. This is given by [15]

$$\frac{dE}{dx} = -[0.97\,\text{MeV/cm}] \times \left(\frac{1}{\beta^2}\ln\left\{\frac{\beta(E+mc^2)}{I}\left(\frac{E}{mc^2}\right)^{1/2}\right\} - 1/2\right). \quad (3)$$

Here, the geometric mean ionization and excitation potential $I = 635$ eV for PbF_2 [16]. The average number of Cherenkov photons created per 511 keV photoelectron is given by

$$N = \int_{511}^{81}\left(\frac{dN}{dx}\right)\left(\frac{dx}{dE}\right)dE. \quad (4)$$

We have evaluated Eq. (4) numerically using an average value of $n^2 = 3$ [9]. In this way, we estimate that around 5 photons will be emitted between 250 and 500 nm per 511 keV γ-ray. Thus, with a 50% photoelectron yield (and neglecting the Compton electrons), a 25% quantum efficiency for the photocathode, and we should expect something like 0.6 photoelectrons per 511 γ-ray. This is orders of magnitude lower than scintillator materials, and under most circumstances would rule this out as a viable detector for positron studies. In this application, however, the detector can be used, although since there is less than one photoelectron per γ-ray, counting statistics will not be optimized. Nevertheless, since PMT saturation can so easily occur in this type of measurement the trade off is ultimately beneficial.

4. Timing measurements

When coupled to an ultra-fast photomultiplier, a Cherenkov radiator can be used for measuring the time width of intense sub nanosecond positron pulses, even if the detection efficiency of such a system is low. The fastest photomultiplier we have found that is suitable for such a measurement is the Hamamatsu model R3809U-50. Its multialkali photocathode has a peak quantum efficiency of 20%. However, this is a two-microchannel plate device, with a thin film covering the input side of the first plate to reduce the sensitivity to ions and eliminate ion feedback. As a consequence of the film, the effective photoelectron collection efficiency is only about 50%. We have obtained a version of this tube with only a single microchannel plate, and no film for reducing ion feedback.

We have used this to monitor the time width of our positron beam, with an 11×11 mm cylindrical PbF_2 crystal, the size of which is determined by the size of the photocathode. The single photon response, as measured using a ^{22}Na test source, is ~260 ps. Although this is likely to broaden somewhat when a large pulse is detected, it is quite sufficient for monitoring our ~1 ns wide pulses. Fig. 1 shows the measured single-electron response as well as a typical bunched positron pulse. The latter was measured as described in Ref. [4]. The small size of the photocathode, and hence of the useful PbF_2 crystal, however, makes this detector unsuitable for SSPALS measurements. Using a larger crystal connected to the small cathode via a light guide could increase the overall detection efficiency, but at the expense of the time resolution. If a 50 mm diameter × 40 mm length crystal were attached to an 11 mm diameter cathode by a light guide then the probability of a

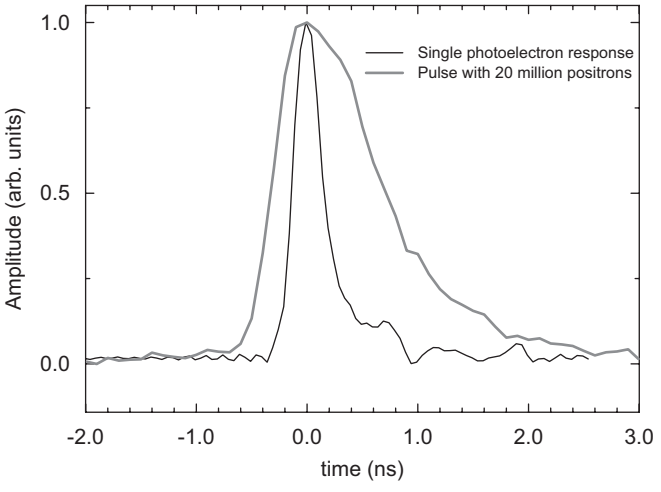

Fig. 1. The single photoelectron response and a pulse containing ~20 million positrons for a PbF_2 Cherenkov radiator coupled to a Hamamatsu R3809U-50 single-channel plate photomultiplier. The single photoelectron response was measured using a small 22Na test source and is an average of 4096 events. Both curves have been normalized to unit peak height. The peak magnitude of the single electron trace was ~50 mV, and that for the pulsed beam was ~120 mV. The data were recorded using an Agilent 54855A digital storage oscilloscope.

photon entering the cathode would be approximately equal to the ratio of the cathode area to the internal surface area of the crystal, multiplied by an exponential decay related to the internal reflectivity of the crystal (neglecting the area of the light guide). That is it would be proportional to around 0.3% per reflection. If each light bounce takes ~0.2 ns then collecting just 5% of the available light would give a time response similar to that obtained using an ordinary PMT.

5. SSPALS measurements

For the SSPALS measurements, a Hamamatsu H3378-50 photomultiplier coupled to a 50 mm diameter × 40 mm length PbF_2 crystal was used. A comparison of this detector and a PWO_4 scintillator attached to a Phillips XP 2020 photomultiplier is shown in Fig. 2. In the former case, the time response is determined entirely by the PMT response and the light collection from the crystal, and is around 3.5 ns FWHM. The latter case is just the opposite; the response is determined almost entirely by the decay time of the scintillator, which is around 15 ns. The anode signal from the PMT is split and sent into an Agilent 54885A digital storage oscilloscope. One channel is recorded at high gain (typically 100 mV/division) and another at low gain (typically 5 mV/division), so that the low level part of the signal can be recorded with minimal noise from the amplifier. These signals are terminated into 50 Ω in the oscilloscope. The two signals are then spliced together to form lifetime spectra of the type shown in Figs. 2 and 4.

One problem that is exacerbated when using a Cherenkov radiator is that the detection efficiency is strongly dependent on the energy of the γ-ray, with lower energy photons detected *less* efficiently. This is in contrast to a scintillator in which lower-energy photons are detected *more* efficiently because they are more likely to stop in the medium. When positrons annihilate directly with electrons two photons, each of energy 511 keV, are created. However, it is also possible for an electron and a positron to form a bound state, known as positronium (Ps) [17]. Ps can exist in a singlet state, called para positronium (*p*-Ps), or a triplet state, called *o*-Ps. For *o*-Ps the electron and positron spins are parallel, and *o*-Ps therefore decays primarily into an odd number of annihilation photons (since to do otherwise would not conserve angular momentum) [18]. Thus, most of the photons arising from *o*-Ps decay have an energy that is less than 511 keV, and are therefore less likely to be detected via a Cherenkov radiator. This means that the amount of Ps present will be underestimated. By considering the *o*-Ps annihilation photon energy spectrum [7], we estimate that approximately one-third of *o*-Ps annihilation events would not be detected because of this. Similarly, one must remain mindful that attenuation may have a larger effect on the detection efficiency than is the case for scintillator-based detectors.

Nevertheless, Cherenkov radiation can still be used for SSPALS measurements. We have tested this by looking at Ps formed on the surface of an Al(1 1 1) single crystal. Positrons implanted into such crystals may diffuse back to

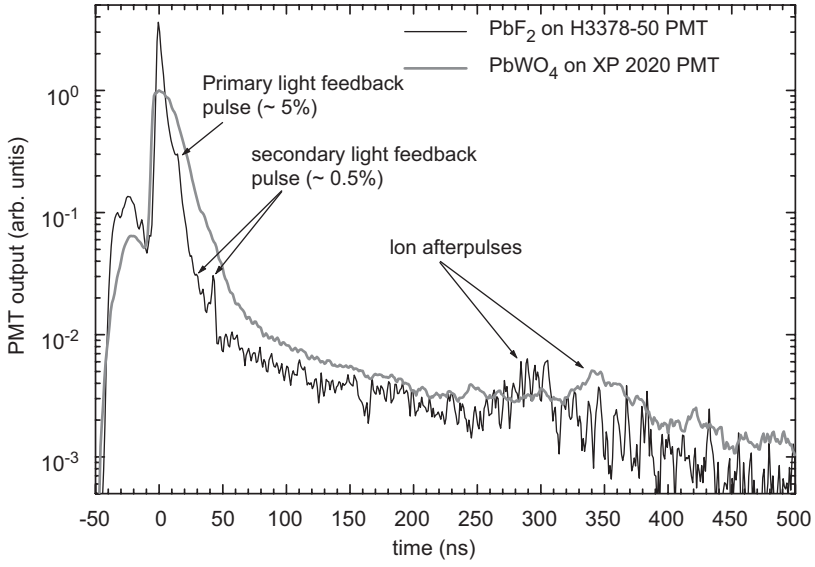

Fig. 2. SSPALS spectra recorded using a PbF_2 crystal attached to a H3378-50 PMT and a PWO_4 crystal on an XP2020 PMT. The positron pulse in each case contains ~8 million positrons with a time width of ~1 ns FWHM and were implanted into an Al (1 1 1) single crystal at ~1.5 keV. The data are normalized to have equal areas. The features present at ~300 and 350 ns are ion after pulses. These are due to residual gas atoms in the tube or adsorbed on dynode surfaces being ionized and accelerated into the cathode and creating a secondary pulse. This occurs in all photomultipliers to some degree. The feature at ~15 ns in the H3378-50 PMT is not present in all phototubes. The manufacturer suggested that it is caused by photoelectrons elastically scattered from some electrode surface that return to the photocathode and are subsequently collected by the first dynode. This process should take much longer than the electron transit time of 15 ns. It is more likely that the after pulse is caused by a small amount of light from the large pulse of electrons crashing into the anode being picked up by the photocathode, thus giving rise to after pulsing that repeats at intervals of almost exactly one transit time. The light feedback pulses are indicated in the figure with their approximate amplitudes, given as a percentage of the primary pulse amplitude.

the surface where some of them are emitted as Ps atoms. In addition to this, positrons trapped in a surface state may be thermally desorbed as Ps atoms [19]. Fig. 3 shows the position of the sample relative to surrounding accelerator electrodes. Ps atoms emitted from the Al surface may collide with these electrodes, which leads to a measured *o*-Ps lifetime (~130 ns) that is less than the vacuum lifetime (142 ns).

Fig. 4 shows lifetime spectra taken at various temperatures; the increase in Ps formation due to thermal desorption is quite apparent. This demonstrates that Ps formation may be measured using a Cherenkov radiator, despite the diminished detection efficiency associated with three photon annihilation.

6. Conclusion

We have shown that the Cherenkov light from a PbF_2 crystal may be used to detect the annihilation radiation produced by an intense burst of positrons. This is an inefficient way to detect low-energy γ-rays, but the technique of SSPALS actually requires a low efficiency of light generation, and this type of detector is certainly not applicable to general positron studies. However, it does

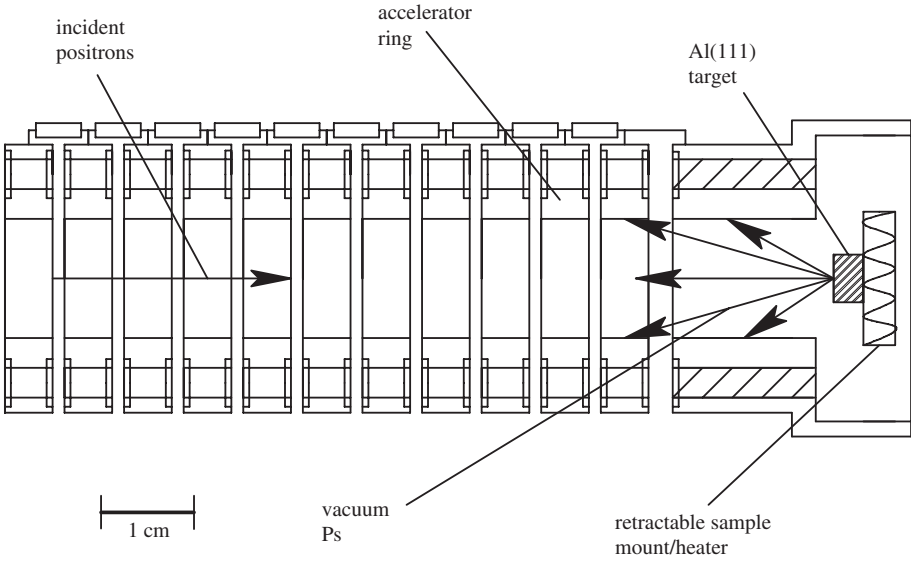

Fig. 3. The accelerator electrodes and position of the Al (1 1 1) crystal. Ps created at the Al surface is emitted into the vacuum and may collide with the surrounding electrodes before self-annihilation occurs, resulting in a reduction in the measured Ps lifetime.

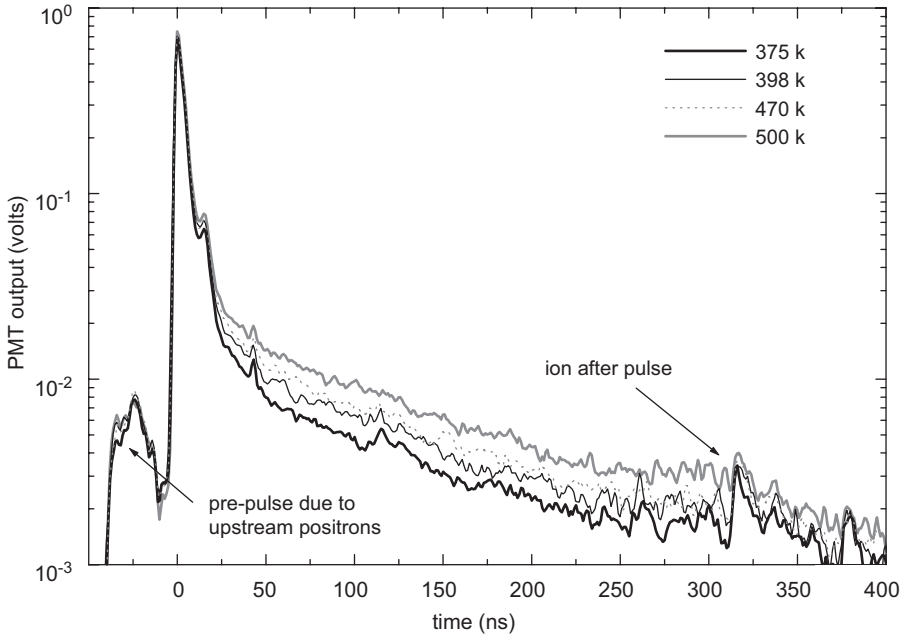

Fig. 4. The increase in positronium formation and the long-lived component of the lifetime spectrum following heating of an Al (1 1 1) crystal is clearly observed using a PbF_2 radiator. The pre-pulse is due to positrons annihilating upstream before the beam reaches the buncher.

provide an excellent way to monitor the time width of a sub-nanosecond positron pulse. Indeed, the main advantage to using a Cherenkov radiator (perhaps the only advantage when low-energy photons are to be detected) is the time response. Cherenkov light is emitted instantaneously, so only scintillators that have decay times shorter than the characteristic time response of the PMT can provide comparable timing signals. The only scintillators that meet this requirement are plastics, which have an extremely low stopping power, even for low-energy γ-rays. Thus, we conclude that a high-density Cherenkov radiator can be usefully applied for the measurement of intense bursts of 511 keV γ-rays, and provides the best possible timing resolution for the technique of SSPALS.

This type of detector may be useful in experiments on the formation of the dipositronium molecule (Ps_2) [20]. If two *o*-Ps atoms form such a molecule they will rapidly annihilate, which means that the amount of long-lived Ps will be reduced when the incident positron beam density is increased [21]. The sensitivity to what are likely to be very small changes in the lifetime spectra (of the order of 1%) is greatly enhanced by the ability to analyze spectra up to early times and so, despite the reduced detection efficiency, a Cherenkov radiator may be the best way to study this effect. Moreover, without such timing information one cannot determine any direct information regarding the rate at which such a process occurs since it is very fast and diminishes with the Ps density. Work is currently underway to perform just this measurement.

Acknowledgments

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