Branching-ratio measurements of multiphoton decays of positronium

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Positronium (Ps) decay to four- and five-photon final states has been measured. From a sample of 3.73×10^{9} detected events of Ps annihilation, the QED-allowed branching ratios of para-Ps to four photons $R_4=1.14(33)(21)\times10^{-6}$ (stat., sys., 68% C.L.), and ortho-Ps to five photons $R_5=1.67(99)(37)\times10^{-6}$ (68% C.L.) were measured. No charge-conjugation symmetry violating events were observed, establishing a branching ratio limit for (o-Ps$\rightarrow4\gamma$) of $R_4^{\not C}<3.7\times10^{-6}$ (90% C.L.) and a new limit for (p-Ps$\rightarrow5\gamma$) of $R_5^{\not C}<2.7\times10^{-7}$ (90% C.L.). The experiment was performed using the Gammasphere array of Compton-suppressed high-purity germanium detectors. Gammasphere's segmentation, high efficiency, and excellent energy resolution allowed event discrimination sufficient to observe the rare decay modes in the presence of substantial backgrounds.

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I. INTRODUCTION

Positronium (Ps) is the bound state of an electron and a positron, and as in ordinary atoms, the physics of Ps is dominated by quantum electrodynamics. While tests of QED in ordinary atoms can yield precise measurements of α, measurements of annihilation lifetimes and branching ratios of Ps test QED calculations to high orders of α. But unlike ordinary atoms, Ps is an eigenstate of the charge-conjugation operator, **C**. For Ps in a state with orbital angular momentum L and total spin S, the **C** eigenvalue is $C_{\mathrm{Ps}}=(-1)^{L+S}$. The photon is intrinsically **C** odd, so the eigenvalue of an n-photon state is $C_\gamma=(-1)^n$. The assumption of **C** symmetry conservation provides a selection rule for annihilation radiation from a state of Ps: $C_{\mathrm{Ps}}=C_\gamma$ implies n photons such that $(-1)^{L+S}=(-1)^n$ [1]. Tests for **C** violation in Ps search for decays to the "wrong" (**C** forbidden) numbers of photons. Measurements of Ps annihilation use the two lowest energy states of Ps (both with $L=0$): the ground state $^1S_0$ parapositronium (p-Ps) and $^3S_1$ orthopositronium (o-Ps). The mean decay lifetime of p-Ps is ≈125 ps, while the lifetime of o-Ps is ≈142 ns. Because the dominant decay mode of p-Ps is two-photon decay, while for o-Ps it is three-photon decay, previous measurements to test **C** in the annihilation of Ps search for the decays to four photons from o-Ps. This requires preparing a sample of Ps, allowing the p-Ps component to decay, and observing the remaining o-Ps for forbidden four-photon decays as distinguished from background events. The result is a limit on a **C**-violating branching ratio such as $R_4^{\not C}\equiv\Gamma(o\text{-Ps}\rightarrow4\gamma)/\Gamma(o\text{-Ps}\rightarrow3\gamma)$. Measurements to test the QED calculated **C**-allowed branching ratios $R_4\equiv\Gamma(p\text{-Ps}\rightarrow4\gamma)/\Gamma(p\text{-Ps}\rightarrow2\gamma)$ and $R_5\equiv\Gamma(o\text{-Ps}\rightarrow5\gamma)/\Gamma(o\text{-Ps}\rightarrow3\gamma)$ are made with a similar technique. These four- and five-photon decay measurements test QED at order α^7 and α^8, but the calculations do not yet require evaluating multiloop diagrams.

II. PREVIOUS WORK ON FOUR- AND FIVE-PHOTON DECAY

The most precise measurement of the **C**-allowed four-photon decay mode branching ratio is $R_4=1.50(11)\times10^{-6}$ [2], compared to the calculated QED value (with one-loop corrections) of $R_4=1.4388(21)\times10^{-6}$ [3]. The first measurement of this rare Ps decay mode found $R_4=1.30(31)\times10^{-6}$ [4]. The four-photon annihilation rate can be written as $\Gamma(4\gamma)=\Gamma_{\mathrm{LO}}(4\gamma)[1-14.5(6)\alpha/\pi+O(\alpha^2)]$, where the lowest order rate is $\Gamma_{\mathrm{LO}}(4\gamma)=0.013\,895\,7(4)m\alpha^7$ [3]. Matsumoto *et al.* [5] found a single five-photon decay event, establishing a branching ratio of $R_5=2.2^{+2.6}_{-1.6}\pm0.5\times10^{-6}$, compared with their tree-level calculation of $R_5=0.9591(8)\times10^{-6}$. Earlier (tree-level) calculations gave $R_5=1.00(6)\times10^{-6}$ [6] and $R_5=0.96\times10^{-6}$ [7]. At tree level, the decay rate to five photons is $\Gamma(5\gamma)=\Gamma_{\mathrm{LO}}(3\gamma)\times[0.0189(11)\alpha^2]$, where the three-photon decay rate is $\Gamma_{\mathrm{LO}}(3\gamma)=2/(9\pi)(\pi^2-9)m\alpha^6$, yielding $\Gamma(5\gamma)\propto\alpha^8$ at tree level [6].

The current limits on **C**-violating decays of Ps are $\Gamma(p\text{-Ps}\rightarrow3\gamma)/\Gamma(p\text{-Ps}\rightarrow2\gamma)<2.8\times10^{-6}$ (68% C.L.) [8], and $\Gamma(o\text{-Ps}\rightarrow4\gamma)/\Gamma(o\text{-Ps}\rightarrow3\gamma)<2.6\times10^{-6}$ (90% C.L.) [9]. The other stringent limits on **C** violation come from the search for ($\pi_0\rightarrow3\gamma$), with an upper limit on the branching ratio of 3.1×10^{-8} (90% C.L.) [10], and ($\eta\rightarrow\pi^0+\mu^+ +\mu^-$) [11]. Our experiment has measured the branching ratios R_4 and R_5 and searched for **C**-violating decays of (o-Ps$\rightarrow4\gamma$) and (p-Ps$\rightarrow5\gamma$) (the first search for this decay mode), deriving limits for $R_4^{\not C}$ and $R_5^{\not C}$.

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III. EXPERIMENT

The present experiment was performed using the Gammasphere array at the 88'' Cyclotron at Lawrence Berkeley National Laboratory [12]. Previous measurements of multiphoton Ps decays used NaI detector arrays [2,4,5,9], and Gammasphere has much better energy resolution, a significant advantage in background suppression in searching for rare multiphoton Ps decays. Background events arise from photons from Compton scattering, bremsstrahlung, and nuclear transition γ rays. Good energy resolution reduces accidental summing of photon energies into an apparent 1022-keV event energy sum characteristic of annihilation. Gammasphere is made up of 110 Compton-suppressed high-purity germanium (HPGe) detectors, which occupy 110 hexagonal surfaces of a 122-element polyhedron surrounding an 18-cm-radius target chamber. Hundred detectors were functioning during this experiment. The empty pentagonal surfaces of the array are often used for additional detectors, such as the β detector in the present work. Each Ge detector assembly consists of a $\sim$7 cm diameter$\times$8 cm long cylindrical HPGe detector surrounded by six bismuth germanate (BGO) scintillators on the sides and one BGO scintillator at the back. Using the BGO detectors in anticoincidence improves the ratio of photopeak to Compton-scattered events. The BGO detectors were shielded from direct radiation by Heavimet (90% tungsten alloy) collimators. The Ge detector faces are 25 cm from the source at the center of the array, subtending a solid angle of 0.5% of 4π. The probability that a detected 511-keV photon deposits its full energy is roughly 15%. The photon energy resolution is well characterized by the relation ΔE (full width at half maximum) $=1.5$ keV $+1.0$ (keV/Mev)$\times E$(MeV), which gives $\Delta E=2.0$ keV at $E=0.5$ MeV. Previous NaI experiments as [2,4,5,9], used customized lead collimator inserts inside the arrays to shield detectors from Compton-scattered events, in which a photon scatters in one detector element, depositing partial energy, and is then scattered back into the array, leaving some or all of its energy in a different detector in a backward direction. Although the Gammasphere has some collimation provided by the Heavimet shields in front of the BGO's (each Ge detector is shielded from backward scattered photons from about 25% of the entire array by the Heavimet in front of the detector module), such scattered events are suppressed in the Gammasphere by cuts on the energy sum and the vector momentum sum of the detector hits in an event. The angular segmentation and the good energy resolution allow distinction between scattered events from full-energy deposition events.

The source of positronium was made with a 10-μCi ^{68}Ge source between two silica aerogel hemispheres. Ps-annihilation experiments usually use either ^{22}Na or (^{68}Ge-^{68}Ga) for positron sources. ^{22}Na has a lower β^+ end-point energy (0.54 MeV, compared to 1.90 MeV for ^{68}Ga), requiring less material to stop the positrons. The 1275-keV transition photon from the daughter ^{22}Ne present in nearly 100% of the β^+ decays is an additional source of backgrounds, but this photon can act as a convenient timing start signal for Ps decay. ^{68}Ge decays with a 288 day half-life by electron capture to ^{68}Ga. ^{68}Ga, while it has higher end-point energy, requiring more stopping material, is a cleaner source of positrons. A γ ray at 1077 keV is present in only 1.3% of the β^+ decays. The timing signal required to distinguish *o*-Ps from *p*-Ps can be obtained by detecting the positron directly in the case of ^{68}Ga.

To search for **C**-violating four-photon decays of *o*-Ps or the rare, but allowed five-photon decays, it is desirable to form as much *o*-Ps as possible. Silica aerogel has been shown to be an efficient production medium [13]. The aerogel sphere had a density of 0.3 g/cm^3, and a diameter of 58 mm. A 2-mm-thick plastic shell, 63 mm in diameter surrounded the aerogel. The mean range of the β^+'s from the ^{68}Ga in the aerogel was calculated using electron gamma shower (EGS) software [15] to be 0.75 cm. EGS predicts that 99.9% of the positrons are stopped within the aerogel. The 10-μCi source, a 3-mm-diameter spot between two 1 mg/cm^2 Kapton foils, was obtained from Isotope Products Laboratory. The ^{68}Ge source was placed between two pieces of 0.2-mm-thick plastic scintillator. The positron detector consisted of the two pieces of scintillator coupled by a 10 cm light guide to a Hamamatsu R1450 photomultiplier tube (PMT). The source and β detector were inserted into the center of the Gammasphere array through an access port and supported by a flange mounted on the Gammasphere target chamber. The EGS software predicted that 90% of the β's from the source, which pass into the aerogel to form positronium give a start signal in the PMT. This figure was consistent with the observed rate in the positron detector (3.6×10^5 Hz) and the source activity. Other studies of Ps formation [13,14] suggest that, of the positrons which enter the aerogel, roughly 30% thermalize and form Ps, while the remainder thermalize and directly annihilate with electrons at rest without forming Ps. The free annihilation of e^+ and e^- at rest in either singlet or triplet *s*-wave initial states is identical for this study to the *p*-Ps or *o*-Ps annihilation process. The free annihilation takes place as soon as the positrons thermalize, less than 1 ns after emission. Of the Ps which is formed in the aerogel, 3/4 begins as *o*-Ps and 1/4 as *p*-Ps. The *o*-Ps is subject to pick-off losses in the source, which shortens the observed *o*-Ps lifetime compared to the predicted mean lifetime of approximately 142 ns. The target chamber was evacuated to 10^{-6} torr to remove oxygen from the aerogel to minimize the pick-off loss rate.

The experiment used a two-level trigger to detect Ps decay events. The first-level Gammasphere trigger is based on a fast linear signal from the Ge detectors. An adjustable discriminator level sets the minimum number of hits in the array required within a 1-μs time window to trigger an event acquisition. The trigger condition was set to two or more Ge detector hits. The main trigger was the presence of a Gammasphere pretrigger, a trigger from the β detector PMT within the 1-μs Gammasphere timing window, and the readout of two or more Ge hits, in which the Compton-suppressing BGO detectors did not fire (''clean'' Ge hits). With the external timing trigger from the β detector with the ^{68}Ge source, detector-dependent timing offsets in the first-level trigger are canceled. A time to amplitude converter

(TAC) for each Ge detector records the time between the β detector PMT pulse and the Gammasphere detector hits. The timing resolution of the setup was determined from events with a valid β detector hit, and two photons detected by Gammasphere, one of which was 511 keV in an arbitrarily chosen reference detector. The Gaussian width of the TAC timing peak with respect to this reference counter was parametrized by $\sigma^T=(179/\sqrt{E})-(448/E)-3.25$, where E is in keV and σ^T is in nanoseconds. At $E=511$ keV, $\sigma^T=4.6$ ns, and at $E=100$ keV, $\sigma^T=11.0$ ns. The energy deposited, the TAC value, and the detector identifier are recorded for each hit detector. The data were written to tape for off-line analysis. The 15-day run had a lifetime of 9.45(1) $\times 10^5$ s, compared to a lifetime of 2×10^7 s in the experiment in Refs. [5,9]. Approximately 1.18×10^{10} events were written to tape (0.8 TB of data) at an acquisition rate of 12.4 kHz. A total of 3.73×10^9 qualify as Ps-annihilation events. The data were compressed and sorted in a preliminary pass in which two-photon annihilation events were counted and events with $n\geqslant 3$ which passed a simple sum-energy cut were saved for later analysis.

IV. DATA REDUCTION

The object of the data reduction is to distinguish real four- and five-photon annihilation events from backgrounds. The number of (p-Ps$\rightarrow 2\gamma$) and (o-Ps$\rightarrow 3\gamma$) events are counted for determining branching ratios. All potential Ps-annihilation events must pass three basic cuts.

(1) The sum energy of the γ's in the event must pass $|\Sigma_{i=1}^{n}E_i-1022\text{ keV}|<5.3$ keV (16 analog-to-digital converter channels).

(2) The TAC value (time between β hit and Gammasphere pretrigger) must be in a 448-ns acceptance window to avoid pileup at the end of the β-PMT trigger window

(3) The photons in the event must have arrival times within one standard deviation of the weighted average time of the event. That is, the residual time of an event, R_T must satisfy

$$R_T=\sqrt{\frac{1}{n}\sum_{i=1}^{n}\frac{(t_i-\langle t\rangle)}{\sigma_i^T}}<1.0, \qquad (1)$$

where $\langle t\rangle$ is the weighted mean time of the event and σ_i^T is the time resolution of the ith photon in the event.
The width of the energy and timing cuts were chosen to maximize the number of accepted real four- and five-photon annihilations, while minimizing the number of accidental background events. The width of the TAC acceptance window (448 ns) is an artifact of the electronics setup requiring an overlap of the PMT trigger and Gammasphere main trigger. We note that the sum energy cut window (± 5.3 keV) is much more restrictive than in the experiments in Refs. [2,5] using Na I arrays.

Candidate Ps events must also pass geometric cuts. Two-photon candidate events must involve two detectors exactly 180° apart in the Gammasphere. The source of positronium is 5.8 cm in diameter and 511-keV photons from off-center decays cannot hit detectors less than 180° apart unless the photons scatter, because the Ge detectors are 7 cm in diameter. To reduce backgrounds from two-photon events in which one or both photons scatter out of one detector and hit another, events with $n\geqslant 3$ must contain no two detectors that are 180° apart. Real (o-Ps$\rightarrow 3\gamma$) events have all three photon momenta in a plane containing the source. For three-photon candidate events, we require that the distance, D to the origin (the center of the Gammasphere array) from the plane containing the three hit detectors be less than the radius of the aerogel (2.9 cm). To reduce (o-Ps$\rightarrow 3\gamma$) generated backgrounds in $n=4$ sample, the distance to the origin from the plane containing any three hits is required to be greater than 2.9 cm. For the $n=5$ sample, any event which has three hits coplanar with the origin ($D=0$) is rejected. The different criteria for D for $n=4$ versus $n=5$ events optimize the discrimination against background events. With five detectory hits, there is a higher probability to obtain a smaller minimum distance D among any three detectors than with a four-photon event. Five-photon background events are more strongly peaked at $D=0$ than are simulated real five-photon decay events, and the allowed kinematic distribution of D for real five-photon decays is more strongly peaked at $D=0$ than for the four-photon decays.

Finally, a cut is made on the vector momentum sum of the event. For $n=2$ events, the momentum sum distribution is dominated by the energy resolution of the Ge detectors and not the granularity of the Gammasphere, and the colinearity requirement is effectively a momentum sum cut at $|\Sigma\vec{p}_i|<30$ keV/c. For $n\geqslant 3$ events, finite source and detector solid angle effects dominate the momentum sum distribution, and cuts for the momentum sum (and D) are optimized with simulated signal and background events. For $n=3$ events, the final momentum sum cut is $|\Sigma\vec{p}_i|<150$ keV/c, while for $n=4$ and $n=5|\Sigma\vec{p}_i|<100$ keV/c. The events surviving these cuts are illustrated in Fig. 1 and enumerated in Table I.

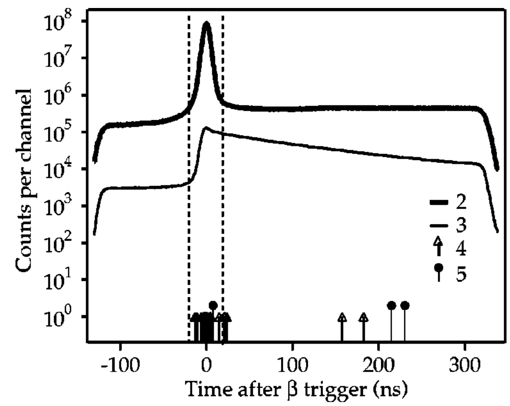

FIG. 1. Ps-annihilation events passing all cuts as a function of time after the β decay ($t=0$). The dashed lines are at ± 20 ns. 78% of the 3γ events have $t>20$ ns, and 83% of the 2γ events occur within $-20<t<20$ ns. The TAC scaling for the horizontal axis is ≈ 244 ps/channel. Five-photon counts have been offset for visibility.

TABLE I. Counts passing all cuts of each n-photon decay mode observed in time bins $-20<t<20$ ns and $t>20$ ns. Some two-photon decays occurring after $t>20$ ns are accidental: events with a β detector timing trigger from one positron for which all photons are missed, and a subsequent (p-Ps$\rightarrow 2\gamma$) decay in the TAC window.

n	$-20<t<20$ ns	$20<t<315$ ns
2	3.0101×10^9	5.3114×10^9
3	1.0907×10^7	4.4302×10^7
4	16	4
5	1	2

The effects of the cuts on the data is shown in Table II, compared to the efficiency of the cuts on simulated decay events.

V. SIMULATIONS

Determining branching ratios for four- and five-photon decay requires knowledge of the multiple photon detection efficiency $\varepsilon(n\gamma)$ of Gammasphere. These detection efficiencies were determined by using a standard simulation of Gammasphere adapted to Ps-annihilation photons. The Gammasphere simulation software [16] (based on GEANT3.2) accounts for active and passive regions in the Gammasphere, the source geometry and the composition, detector energy thresholds, energy resolution, and any nonfunctioning or missing detectors during the run. Identical cuts were applied to the simulated and real data. The point of origin of the Ps-annihilation photons is randomized to agree with the ^{68}Ge positron range distribution obtained from the EGS simulation. Tests of possible displacements of the Ps source from the geometric center of Gammasphere were performed in the simulations to estimate position-dependent systematic uncertainties in $\varepsilon(n\gamma)$. No statistically resolved change in $\varepsilon(n\gamma)$ was found for displacements less than 1 cm from the center of Gammasphere.

TABLE II. Events remaining after each cut is made on the data for four- and five-photon events. The number of simulated events passing the cuts is also given. The timing cut is not present in the Monte Carlo generated events: all detector hits in the simulation are assumed to be simultaneous. The efficiency $\varepsilon(4,5\gamma)$ is then corrected for the acceptance cut of $\langle t\rangle\pm1\sigma$. NA denotes not available.

Cut	Data	Simulated
$n=4$		
Input events	n/a	110575
Detect $n=4$	2.60187×10^8	577
$\Sigma E=1022$ keV	143977	210
Prompt timing	67583	NA
No colinearity	45117	179
Planarity ($D>2.9$ cm)	70	30
$\lvert\Sigma\vec{p}\rvert<100$ keV/c	20	29
$n=5$		
Input events	n/a	120000
Detect $n=5$	3.04452×10^7	168
$\Sigma E=1022$ keV	859	61
Prompt timing	109	NA
No colinearity	71	49
Planarity ($D>0.0$ cm)	10	35
$\lvert\Sigma\vec{p}\rvert<100$ keV/c	3	32

Different event generating softwares are required for different numbers of final-state annihilation photons. Two-photon annihilations are simple, requiring only two randomly generated angles, θ and ϕ. Three-photon annihilation events were generated using a recursive algorithm described in Ref. [17], which produces two correlated angles (in a plane) between two of the three photons according to the physics of the (o-Ps$\rightarrow 3\gamma$) decay, and then generating Euler angles to randomize the orientation of the decay plane in Gammasphere. Four- and five-photon decay events were produced using the GRACE/BASES/SPRING software package [18]. This software generates all tree-level Feynman diagrams for a QED process and calculates matrix elements by Monte Carlo integration of the graphs. After the phase-space integration, the package SPRING can be used to generate events with probability weighted by the allowed phase space in the integration. The software requires as input an appropriate description of the kinematic phase space for the process. The kinematic description encodes the transformation from dummy integration variables to kinematic variables. For four-photon decay, eight integration variables are required (of which three are trivial Euler angles) as described in the calculation of R_4 in Ref. [3], and for five-photon decay, 11 variables are required [6]. Generated events from this process were used in the GEANT simulation to determine $\varepsilon(4\gamma)$ and $\varepsilon(5\gamma)$. The output of the event generator for (p-Ps $\rightarrow 4\gamma$) was checked against previous calculations of energy and angular distributions [2,7], and the energy spectrum of the generated (o-Ps$\rightarrow 5\gamma$) checked with that calculated in Ref. [7]. No disagreements in the angular or energy spectra were found beyond the statistical uncertainty of the event generation.

The calculated Gammasphere detection efficiency for (p-Ps$\rightarrow 2\gamma$) is $\varepsilon(2\gamma)=3.964(48)\times10^{-2}$, and the efficiency for (o-Ps$\rightarrow 3\gamma$) events is $\varepsilon(3\gamma)=5.66(20)\times10^{-3}$. The errors are statistical, limited by the number of Monte Carlo events. The total number of two- and three-photon decays within the 448-ns TAC window is simply $N(n)/\varepsilon(n)$. In the experiment, we observed $3.640\times10^9 2\gamma$ events and $5.710\times10^7 3\gamma$ events, corresponding to $9.18(11)\times10^{10}$ (p-Ps $\rightarrow 2\gamma$) and $1.010(36)\times10^{10}$ (o-Ps$\rightarrow 3\gamma$) decays. This implies an 11% (o-Ps$\rightarrow 3\gamma$) counting fraction relative to the total number of observed annihilations. The pick-off loss fraction of o-Ps in our source can be estimated by comparing the lifetime of the observed 3γ events to the QED calculated o-Ps lifetime in vacuum of 142 ns. Fitting the 3γ time data in Fig. 1 to a single exponential decay (with a Gaussian time convolution of 12 ns to account for the experimental time resolution), we find the mean lifetime of the o-Ps in the source to be 112.4 ± 1.3 ns. In this fit, we use only data from

$t=+20$ ns to $+310$ ns to avoid any contribution from free positron-electron annihilation. The observed 112 ns lifetime is consistent with other Ps sources using silica aerogel [13,14] and implies a pick-off probability for *o*-Ps of $(1-\tau_{\rm Obs}/\tau_{\rm vac})=21\%$.

For the (p-Ps$\rightarrow 4\gamma$) events, we obtain $\varepsilon(4\gamma)=1.60(30)\times 10^{-4}$ (statistical uncertainty). From the simulations, only 13.8% of the detected four-photon events having the proper sum energy pass the cuts on geometry and momentum (see Table II). For (o-Ps$\rightarrow 5\gamma$), we obtain $\varepsilon(5\gamma)=1.63(29)\times 10^{-4}$ (statistical uncertainty). A 32% correction is applied to the efficiencies $\varepsilon(4\gamma)$ and $\varepsilon(5\gamma)$ to account for the timing cut on R_T, which would reject some real events occurring outside the 1σ time window. For **C**-violating (o-Ps$\rightarrow 4\gamma$) or (p-Ps$\rightarrow 5\gamma$), the detection efficiencies are not necessarily the same as $\varepsilon(4\gamma)$ and $\varepsilon(5\gamma)$, because the event topologies are different for the **C**-violating and **C**-allowed QED decays. Limits on **C**-violating branching ratios from Ps-annihilation experiments are therefore, model dependent. A previous Ps-annihilation experiment addressed this issue by using a **C**-violating QED-like model to provide event topologies, which guided the experiment design [19].

VI. BACKGROUNDS

Several mechanisms generate background four- or five-photon events, which pass the experimental cuts. Reference [4] lists many such potential accidental contributions. The dominant contribution (in four-photon events) comes from bremsstrahlung of the detected positron as it stops in the aerogel and then decays to three photons. If one or more of the annihilation photons deposit only partial energy in a Ge detector, the detected bremsstrahlung photon can move the summed energy and momentum into the accepted ranges. In an EGS calculation, 4.1% of the ^{68}Ga β^+'s produce bremsstrahlung photons with energies higher than the 10 keV detector threshold. Another source of background arises from γ rays from the transition $^{68}{\rm Ga}(1^+)\rightarrow {}^{68}{\rm Zn}(2^+)$ at 1077 keV in 1.3% of the β^+ decays. The Gammasphere detector response to input bremsstrahlung and nuclear γ rays was simulated to produce spectra of detected photons from such events. The other major background source is the accidental summing of a second two- or three-photon Ps decay within the ≈ 16 ns time window. Background contributions from accidentals and bremsstrahlung were estimated using an ''event-mixing'' technique. Using the raw experimental data, we constructed artificial background events by combining two- or three-photon events with a simulated bremsstrahlung, a nuclear γ-ray hit, or a second event. The number of these $(2\gamma+2\gamma)$, $(2\gamma+3\gamma)$, $(3\gamma+3\gamma)$, $(3\gamma+\gamma_{\rm brem})$, etc. events in the experimental dataset was estimated as the product of the square of the source activity, the time acceptance window, the total data acquisition time of the experiment, the probability for a positron to decay by 3γ or 2γ annihilation, the probability for Gammasphere to detect $n=2$ or 3 photons, the probability to emit and detect a bremsstrahlung or transition photon, and the relevant combinatorial factor. The estimated counts from each background mode are enumerated in Table III. The simulated background counts are normalized as the number of event-mixed events passing experimental cuts, multiplied by the ratio of the number of simulated events to the number estimated to be in the dataset.

TABLE III. Sources of background counts for 4γ and 5γ Ps decays. *B*, the number of expected background events is calculated as the product of the absolute number of events of each event type estimated to have occurred during the run, and the efficiency for a simulated event (derived from event mixing) to pass 4γ or 5γ cuts. The uncertainty in each entry is $\pm 30\%$, dominated by the uncertainty in the source activity and the statistical uncertainty from simulations to derive detection probabilities. (Some event types with <0.01 events are omitted in the tabulation but included in the sums.)

Event type	Absolute	*B*	
		$\lvert t\rvert\leqslant 20$ ns	$t>20$ ns
Four-photon decays			
$2\gamma+\gamma_{\rm brem}+\gamma_{\rm brem}$	1.33×10^4	0.016	0.003
$3\gamma+\gamma_{\rm brem}+\gamma_{\rm brem}$	198	<0.05	<0.05
$2\gamma+2\gamma$	2.26×10^6	0.079	0.014
$2\gamma+3\gamma$	1.44×10^6	0.065	0.24
$3\gamma+3\gamma$	9.14×10^5	0.074	0.27
$3\gamma+\gamma_{\rm brem}$	2.23×10^5	1.3	0.01
$3\gamma+\gamma_{1077}$	9.34×10^3	0.34	0.01
$2\gamma+2\gamma+\gamma_{\rm brem}$	9.44×10^4	0.08	0.012
$2\gamma+2\gamma+\gamma_{1077}$	3.95×10^3	0.015	0.002
$\Sigma 4\gamma$		2.1	0.65
Five-photon decays			
$3\gamma+\gamma_{\rm brem}+\gamma_{\rm brem}$	10	0.01	0.01
$2\gamma+3\gamma$	7.99×10^4	<0.03	<0.03
$3\gamma+3\gamma$	1.35×10^5	0.04	0.1
$2\gamma+2\gamma+\gamma_{\rm brem}$	1.88×10^4	0.004	0.004
$2\gamma+2\gamma+\gamma_{1077}$	787	0.001	0.001
$\Sigma 5\gamma$		0.25	0.1

VII. MEASURED BRANCHING RATIOS

To extract the branching ratios, the total number of decays in appropriate time regions shown in Fig. 1 was used. Events in the region between $-20<t<20$ ns come predominantly from *p*-Ps (and free e^+-e^-) decay, while events in the region $20<t<315$ ns come from *o*-Ps decay. We observe that 21% of the total three-photon events and 83% of the two-photon events occur in the first (''short'') time region. The number of events is given in Table I. Subtracting appropriate backgrounds from Table III and using Monte Carlo simulated detection efficiencies, we calculated branching ratios as (for example)

$$R_4=\frac{[N(4,{\rm short})-B(4,{\rm short})]\varepsilon(2\gamma)}{N(2,{\rm short})\varepsilon(4\gamma)}, \qquad (2)$$

where B is the summed background contribution for the appropriate time window. The results (including 90% confidence level limits as in Ref. [20]) for R_4, R_5, $R_4^{\not C}$, and $R_5^{\not C}$ are shown in Table IV. Systematic uncertainty comes from

TABLE IV. Measured branching ratios for four- and five-photon decays of Ps. Errors are statistical, systematic.

Decay mode	1σ errors	90% C.L. limit
p-Ps$\rightarrow 4\gamma$	$1.14\pm 0.33\pm 0.21\times 10^{-6}$	$<1.92\times 10^{-6}$
o-Ps$\rightarrow 5\gamma$	$1.67\pm 0.99\pm 0.37\times 10^{-6}$	$<6.4\times 10^{-6}$
o-Ps$\rightarrow 4\gamma$	$2.0\pm 1.0\pm 7.7\times 10^{-7}$	$<3.7\times 10^{-6}$
p-Ps$\rightarrow 5\gamma$	$0.3\pm 0.3\pm 1.3\times 10^{-7}$	$<2.7\times 10^{-7}$

the uncertainty in $\varepsilon(n\gamma)$. For R_5, the entire timing range of $-20<t<315$ ns was used. This measurement of R_5 improves the previous result given in Ref. [5] and agrees with the QED calculations. The (o-Ps$\rightarrow 5\gamma$) events are expected to follow the time distribution of the (o-Ps$\rightarrow 3\gamma$) events. The normalized likelihood for the observed time distribution of the three five-photon events in Fig. 1 is 78%. That is, given three events occurring at random times [consistent with the distribution of (o-Ps$\rightarrow 3\gamma$) events], 78% of such event triples have a probability less likely than for the three five-photon events in Fig. 1. In the value for $R_4^{\not C}$, 3.11 ± 0.58 background counts from (p-Ps$\rightarrow 4\gamma$) occurring at $t>20$ ns have been subtracted. These events come from pick-off decay of o-Ps to 2γ and accidental (p-Ps$\rightarrow 2\gamma$) decays occurring within the time window. The uncertainty in this background comes from the uncertainty in $\varepsilon(4\gamma)$. Similarly, in $R_5^{\not C}$, 0.4 ± 0.2 events from (o-Ps$\rightarrow 5\gamma$) occurring between $-20<t<20$ ns have been subtracted. The 90% confidence limit for $R_4^{\not C}$ is slightly worse than Ref. [9]: the present experiment observed four potential **C**-violating events with an estimated background of 3.75 events, with an observed sample of ($t>20$ ns) (o-Ps$\rightarrow 3\gamma$)$=N(3\gamma)/\varepsilon(3\gamma)=7.8\times 10^9$ and $\varepsilon(4\gamma)=1.60\times 10^{-4}$, while Yang *et al.* observed three events with an estimated 3.4 background events, and an o-Ps sample of 4.9×10^{10} and $\varepsilon(4\gamma)=3.3\times 10^{-5}$. The new limit on $R_5^{\not C}$ from (p-Ps$\rightarrow 5\gamma$) would relate to more speculative **C**-violating processes than the limit on $R_3^{\not C}$ from (p-Ps$\rightarrow 3\gamma$) as in Ref. [8].

The limiting factor in this experiment was the acquisition time, although past experiments to search for **C**-violating Ps-annihilation modes had total data acquisition times more than a factor of 10 longer. Further Ps-annihilation experiments in Gammasphere could have higher Ps decay detection efficiency. On-line hardware based cuts can be made in the data to prevent readout when the sum energy of an event is less than a user-defined energy threshold. This technique could reduce the readout dead time of the system and could increase the maximum data rate by 40%. The backgrounds in such an experiment could be reduced by roughly a factor of 3 by using a source with less activity (to reduce accidental rates), by better rejection of bremsstrahlung photons in analysis, and by using more complex, detector-dependent timing resolution analysis.

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