

for extensive literature by C. W. F. Everitt *et al.*

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W. B. Dress, J. K. Baird, and N. F. Ramsey, *Phys. Rev. Lett.* **19**, 381 (1967). It is, in principle, possible that for  $\alpha$ 's obtained by the neutron experiments are different from the ones obtained by EFG and HTP.

## Precision Measurement of the Decay Rate of Orthopositronium in SiO<sub>2</sub> Powders\*

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The decay rate of orthopositronium is found to depend linearly on the mass per free volume in powders of SiO<sub>2</sub> with grain radii of 35 and 70 Å. Three data points have a decay rate significantly less than both the theoretical free-space decay rate and the results of previous experiments. The value of  $\lambda_3$  extrapolated to zero density,  $7.104 \pm 0.006 \mu\text{sec}^{-1}$ , is  $(1.9 \pm 0.1)\%$  below the theoretical value of the free-space lifetime. Possible interpretations of these results are discussed.

The measurement of the decay rate of the  $n=1$  <sup>3</sup>S<sub>1</sub> state of positronium (*o*-Ps) provides a clean test of quantum electrodynamics, since to high order the constituent leptons interact only through the electromagnetic interactions. The theoretical value of the decay rate of *o*-Ps including radiative corrections is<sup>1-3</sup>

$$\begin{aligned} \lambda_3^{\text{free}} &= \lambda_3^{\text{free}}(0) [1 + (\alpha/\pi)(1.86 \pm 0.45)] \\ &= 7.242 \pm 0.008 \mu\text{sec}^{-1}, \end{aligned} \quad (1)$$

where

$$\lambda_3^{\text{free}}(0) = \frac{2}{9} \frac{\alpha^6}{\pi} \frac{mc^2}{\hbar} (\pi^2 - 9) = 7.212 \mu\text{sec}^{-1} \quad (2)$$

is the decay rate neglecting radiative corrections.

Previous experiments to determine the free annihilation rate have been performed in gases at pressures ranging from about 1 to 10 atm. The decay rate of *o*-Ps is measured as a function of gas density and the free annihilation rate is interpreted as the decay rate obtained from a linear extrapolation to zero density. This requires an extrapolation of about 5% in  $\lambda_3$ . The results are  $7.262 \pm 0.015 \mu\text{sec}^{-1}$ <sup>4</sup> and  $7.275 \pm 0.015 \mu\text{sec}^{-1}$ .<sup>5</sup> Other results have been reported<sup>6</sup> but are not considered to be precision measurements of  $\lambda_3^{\text{free}}$ .<sup>7</sup> Thus, the average experimental value is about  $2\frac{1}{2}$  standard deviations above theory.

We have attempted to determine the free annihilation rate in a low-density finely divided powder instead of a gas. The compact size (2 in. diameter) of the powder target permits a high  $\gamma$  detection efficiency which reduces noise and also

allows, for the first time, a direct measurement of  $\lambda_3$  by detecting all three of the annihilation  $\gamma$  rays in coincidence. In addition, the extrapolation to zero powder density is less than 1% of  $\lambda_3$ . The possibility that such an extrapolation may actually be interpreted as  $\lambda_3^{\text{free}}$  will be discussed at the conclusion of our paper where we present various interpretations of our data. We have selected two samples of silicon dioxide manufactured by Cabot Inc. (Cab-o-sil) which consist of roughly spherical primary particles with average radii of 35 and 70 Å. If Ps is excluded entirely from the powder grains<sup>8,9</sup> and interacts with the grains only during collisions, then the parameter analogous to the gas density is the mass per free volume of the powder. Hence we plot the annihilation rate as a function of the dimensionless variable

$$\rho^* = \rho / (\rho_{\text{solid}} - \rho), \quad (3)$$

which is proportional to the free-volume density of the powder. Here  $\rho_{\text{solid}}$  for SiO<sub>2</sub> is the solid aggregate density of 2.20 g cm<sup>-3</sup>. The value of  $\rho^*$  is changed by compressing the powder over a density range of  $\rho$  of 0.03 to 0.26 g cm<sup>-3</sup>.

The measurements were carried out with a multiple-detector delayed-coincidence  $\gamma$ -ray spectrometer. The system is used to measure the time interval between positron emission and the detection of a predetermined number (1 or 3) of the *o*-Ps annihilation  $\gamma$  rays. The time-interval measurements are made using an Ortec 467 time-to-amplitude converter (TAC) and a Nuclear Da-

ta 2200 multichannel analyzer (MCA). Both the positron and the  $\gamma$ -ray signals must meet a set of energy and fast-coincidence requirements.

The apparatus is shown schematically in Fig. 1. A 0.15- $\mu$ Ci  $\text{Ge}^{68}$  positron source is placed in the center of a molded plastic scintillator cap, 0.9 mm in diameter and 0.5 mm thick. Approximately 70% of the source positrons pass through the scintillator, are detected, strike the powder target, and form Ps with 15% efficiency. The signal from the plastic scintillator is used as the  $t=0$  signal (TAC start) for  $o$ -Ps formation, since formation occurs within  $10^{-10}$  sec of positron emission.

A set of three 4-in.  $\times$  4-in. NaI detectors coupled to RCA 4522 photomultiplier tubes are used to search for the  $o$ -Ps annihilation  $\gamma$  rays in the 1.0- $\mu$ sec interval after a TAC start signal. The three detectors are mounted in a plane at  $120^\circ$  to each other and with each detector subtending a solid angle at the source of  $d\Omega/4\pi = 0.24$ . A valid time interval is recorded if the following requirement is met: Any of the three NaI detectors has detected a  $\gamma$  ray with an energy in the range  $100 < E < 450$  keV and at a time greater than 25 nsec. The TAC start signal is suppressed if any of the three NaI detectors has detected a  $\gamma$  ray in less than 25 nsec. This prompt rejection eliminates noise and unnecessary dead time by rejecting  $2\gamma$  decay events that occur within a few nanoseconds of emission.

The expected shape of the spectrum beyond the suppressed prompt peak is approximately<sup>4</sup>

$$N(t) = [A \exp(-\lambda_3 t) + B] \exp(-\lambda_s t), \quad (4)$$

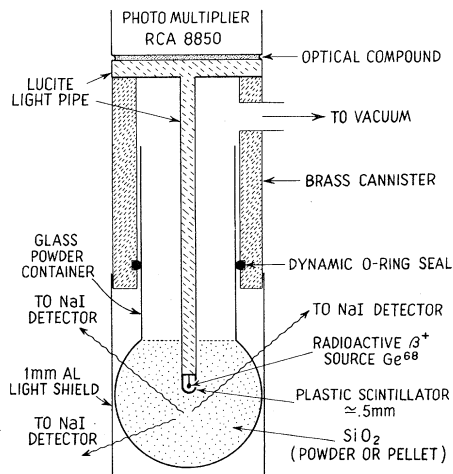


FIG. 1. Evacuated source and powder holder assembly.

where  $\lambda_s$  is the stop rate to the TAC and  $B$  represents the intensity of the uncorrelated background counts. This equation neglects terms in the complete expression<sup>10</sup> which could cause a systematic error in the decay rate determined from our data of at most 0.005%. Furthermore, in this experiment,  $\lambda_s$  is at most about 1000 counts/sec which is about  $10^{-4}$  of  $\lambda_3$  and can therefore be neglected. Thus the spectrum is fitted by a maximum-likelihood program<sup>11</sup> with

$$N(t) = A \exp(-\lambda_3 t) + B, \quad (5)$$

where  $A/B$  is typically 200–400 at the starting point of the fitting program. A slight deviation from the expected shape of the spectrum was noticed through a 0.5–0.6% increase in the lifetime as the starting point of the fitting program was successively stepped out in 33-nsec intervals from 80 to 450 nsec after the prompt peak. The lifetime asymptotically approaches a constant value and the  $\chi^2$  of the fit, which is initially poor, approaches the number of degrees of freedom as the starting point of the fit is stepped out. Sufficiently far from the prompt peak the difference of the fitted spectrum and the data shows no systematic effects. A fit of the data for two exponential components does indicate the presence of a second component with a lifetime of  $45 \pm 5$  nsec and with an intensity 2% that of  $o$ -Ps. The origin of this effect is not understood but it is probably a  $2\gamma$  decay process since no effect was observed in experiments that required the detection of all three annihilation  $\gamma$  rays in coincidence.

Careful consideration has been given to calibrating the time interval of each of the 370 channels (bins) in the spectrum. The TAC-MCA system linearity, or relative bin width, is determined by applying to the TAC a clocked start pulse and a random stop pulse of rate  $\lambda_s = 10^4$  counts/sec. The number of counts in a given time bin is proportional to the width of the bin,  $\Delta t$ , after the appropriate correction factor,  $\exp(\lambda_s t)$ , has been multiplied through. This correction is less than 1% at full scale. A complete time calibration requires that both the linearity and the absolute time between two widely separated points in the spectrum be known. The time width of each bin is then

$$\Delta T_i = n_i T_{jk} \left( \sum_{i=j}^k n_i \right)^{-1}, \quad (6)$$

where  $n_i$  is the number of counts in the  $i$ th bin of the corrected linearity spectrum and  $T_{jk}$  is the time interval between the  $j$ th and  $k$ th bins. An

Ortec 462 time calibrator (TC) was used for daily calibration. The time base of the time calibrator was found to have a systematic error of 20 ppm by comparison with a Hewlett Packard 5345A time digitizer and a Monsanto 3100A frequency synthesizer. In addition to the time calibrator, a Hewlett Packard 8004A pulse generator set to double pulse with a known delay was used to verify the results to 0.1%. The overall time calibration error is less than 0.05%.

The annihilation rates we obtained from our powder samples at various values of  $\rho^*$  are shown in Fig. 2. The following features should be noted: (a) The data are linear and the ratio of the slopes is 1.4. (b) There are three points for which the measured decay rate is lower than the theoretical value. (c) The value of  $\lambda_3$  extrapolated to  $\rho^* = 0$  is in agreement for both sets of data. (d) The average of the two extrapolated values,  $7.104 \pm 0.006 \mu\text{sec}^{-1}$ , is  $(1.9 \pm 0.1)\%$  below the theoretical value and  $(2.3 \pm 0.1)\%$  below the average experimental value obtained in gases.

To verify the accuracy of these results a set of systematic tests was performed. To eliminate any possible effects of count rate or photomultiplier-scintillator dead time the decay rate of the second 70-Å data point was measured using the following four different criteria for acquiring the time spectrum: (1) The usual three NaI detectors were used requiring only one of three anni-

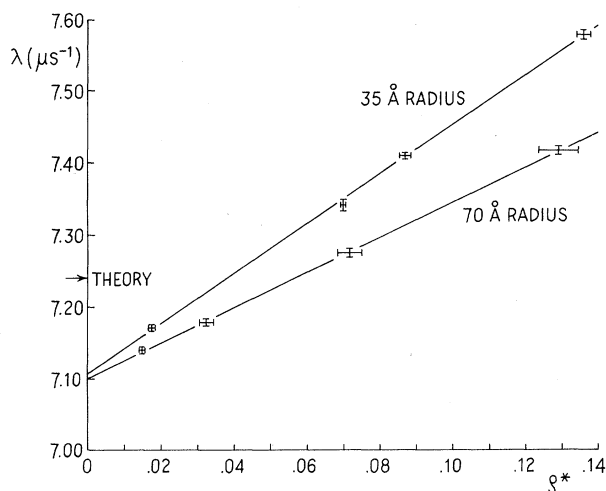


FIG. 2. Plot of decay rate versus  $\rho^*$  for samples of  $\text{SiO}_2$  with either 70- or 35-Å primary particle radius. The  $\rho^* = 0$  intercepts are  $7.101 \pm 0.009$  and  $7.107 \pm 0.008 \mu\text{sec}^{-1}$ . The lowest three points are from powder samples while the rest are from powder pellets. All samples are evacuated to  $10^{-6}$  Torr and outgassed at  $350^\circ\text{C}$  for 4 h.

hilation photons to be detected. Each photon had to have energy in the range  $100 < E < 450 \text{ keV}$ . (2) All three photons had to be detected simultaneously, each with energy in the range  $100 < E < 450 \text{ keV}$ . (3) The three NaI detectors were replaced with a single Pilot-B plastic scintillator 4 in. in diameter by 3 in. thick and it was required that only one of the three annihilation photons be detected. (4) Same as (3) but the prompt-rejection scheme was eliminated so that the entire spectrum was recorded. The results are summarized in Table I. All the measured decay rates agree within experimental error. In addition, two other data points were examined using the  $3\gamma$ -coincidence requirement and yielded results consistent with the single-photon requirement. The  $3\gamma$ -coincidence spectrum had a signal-to-noise ratio ( $A/B$ ) of 2000–3000, a factor of 500 greater than that of any previous measurements in gases.

To check our overall ability to measure a decay rate, the lifetime,  $\tau = \lambda^{-1}$ , of the 14-keV excited nuclear state of  $\text{Fe}^{57}$  was determined and compared with the previous results of  $140.95 \pm 0.3 \text{ nsec}^{12}$  and  $141.25 \pm 0.3 \text{ nsec}^{13}$ . The time interval between the 122-keV  $\gamma$  ray which signals the formation of the 14-keV state and the 14-keV  $\gamma$  ray was measured with our TAC-MCA system using a 4-in.  $\times$  4-in. NaI detector or, as a separate check, a 4-in.  $\times$  2-in. plastic scintillator to detect the 122-keV  $\gamma$  ray. The results are  $141.21 \pm 0.14$  and  $141.24 \pm 0.14 \text{ nsec}$ , respectively, using a  $0.4\text{-}\mu\text{Ci}$  source which yielded start and stop rates to the TAC comparable with those used (1000–2000 counts/sec) in the Ps experiment.

For the density range investigated in  $\text{SiO}_2$ , both the linearity and the dependence of the slope on particle size are consistent with a model in which o-Ps is essentially free in the intergrain space and only interacts with the powder surface during collisions. With this interpretation,  $\lambda_3^{\text{free}}$  is the  $\rho^* = 0$  intercept. Since this strongly contradicts theory and prior experiments, the validity of extrapolating to zero density must be questioned. However, three data points are conclu-

TABLE I. Decay rate ( $\mu\text{sec}^{-1}$ ) of the second 70-Å datum point acquired by four different methods.

1. NaI, single $\gamma$	$7.174 \pm 0.008$
2. NaI, three $\gamma$	$7.180 \pm 0.012$
3. Plastic, no rejection	$7.179 \pm 0.007$
4. Plastic, prompt rejection	$7.181 \pm 0.008$

sively (1–1.5%) below the theoretical value. Several alternate explanations have been considered.

(1) Instrumental error. All the systematic checks including the  $\text{Fe}^{57}$  measurement exclude any reasonable possibility of a 1–2% systematic error.

(2) Deviations from the expected spectrum shape. Long-lived positrons that either decay into two  $\gamma$  rays (possibly the origin of the previously mentioned 45-nsec component) or eventually form  $o$ -Ps may affect the fit. We feel that this is unlikely since the positron lifetime must be longer than that of  $o$ -Ps to explain the observed effect.

(3) Physical effects due to the powder environment. A number of processes related to surface binding at the grains and to electric fields (Stark shift) in the intergrain spaces have been and are being investigated. The result of these considerations is that the data reported above cannot be plausibly explained as due to the powder environment. A detailed theoretical discussion of the effect of such processes will shortly be submitted for publication by Ford, Witten, and Sander.<sup>14</sup>

In summary, we report the discovery of two new effects: (a) linearity of  $\lambda_3$  versus  $\rho^*$  in  $\text{SiO}_2$  powders with  $\lambda_3(\rho^* = 0) = 7.104 \pm 0.006 \mu\text{sec}^{-1}$ ; (b) actual measured values of  $\lambda_3$  below the currently accepted value of  $\lambda_3^{\text{free}}$ . A conclusive understanding of these effects and in particular of the possibility of identifying  $\lambda_3(\rho^* = 0)$  as  $\lambda_3^{\text{free}}$  should follow from further work now underway in our laboratory. Our program includes a study of  $\lambda_3$  versus temperature, attempts at lowering the  $\text{SiO}_2$  density, and work with other oxide powders.

We can also use our apparatus to improve the precision of both the  $\text{Fe}^{57}$  decay rate and the isomeric shift of this decay rate.

We thank Professor G. W. Ford, Professor L. Sander, Professor T. Witten, and Professor W. Brandt for helpful discussions regarding the interpretation of our data. Professor P. Orth kindly supplied the maximum-likelihood fitting subroutine we used in our data analysis.

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