From the definitions of R and $\bar{R}$ it follows immediately that

$$\lambda\bar{R}^{-1}(\lambda+ip)-(\lambda+ip)\bar{R}^{-1}(\lambda-i\epsilon) = -iph+\lambda R^{-1}(\lambda+ip)-(\lambda+ip)R^{-1}(\lambda-i\epsilon) \ . \tag{A16}$$

Substituting this into (A15) yields, after some algebra,

$$\hat{g}(p)=\beta h\,\hat{c}_2(p)+\beta H/4p \ , \tag{A17}$$

where $\hat{c}_2(p)$ is the longitudinal autocorrelation function defined in Sec. IV. Of course, the same result could have been found by noting that we have, by expanding Eq. (2.16) to linear terms in β,

$$\langle S_0^z(t)\rangle = -\beta\,\mathrm{Tr}\mathcal{K}(H+h)S_0^z(t)/\mathrm{Tr}1 \ . \tag{A18}$$

Equation (A18) can also be rewritten as

$$\langle S_0^z(t)\rangle = \beta\,[h\,\mathrm{Tr}S_0^zS_0^z(t)-\mathrm{Tr}\mathcal{K}(H)S_0^z(t)]/\mathrm{Tr}1 \ , \tag{A19}$$

which, indeed, agrees with (A17).

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Correlation between Lifetime and Momentum for Positron Annihilations in Teflon

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A novel technique has made it possible to correlate the three components in the lifetime distribution of positrons in Teflon with the angle between the γ rays resulting from the annihilation. The intensity of the intermediate component was found to be virtually independent of this angle, whereas the intensity of the longest-lived component was greatly enhanced for angles corresponding to higher momentum. Apparently the intermediate component does not result from the annihilation of orthopositronium, because the momentum associated with this component is too small. The intermediate component could be caused by free-positron annihilation or by annihilation of positrons which are bound in Teflon molecules.

INTRODUCTION AND HISTORY OF PROBLEM

As early as 1956 it was suggested[1,2] that many details of positron interactions with matter could be clarified by an experiment in which the positron lifetime was correlated with the angle between the two γ rays resulting from the annihilation. Experimental techniques needed for such a measurement were not developed until several years later. In 1964, preliminary results were reported[3] which showed for the first time the angular dependence of the long-lived component in the lifetime spectrum of positrons in Teflon.

Additional measurements, using some refinements in technique and with improved statistics, were reported by one of us (V. F. W.).[4] These results indicated that the angular correlation of radiation from "pick-off" annihilation of positrons in positronium (Ps) atoms is significantly broader than the angular correlation of annihilation radia-

tion from free positrons, and it was pointed out that this broadening of the angular correlation resulted from the contribution of the orbital motion of the positron to the momentum of the annihilating pair. Thus it appeared that this technique could be quite useful in studying the chemistry of Ps in such materials.

Unfortunately, because of the need to compromise among the conflicting requirements of time resolution, angular resolution, and counting efficiency, these results suffered from rather poor time resolution. In another experiment of this type, [5] a much hotter source was used in conjunction with smaller scintillators, and the efficiency was improved because the sample under study was the scintillator itself; but the resulting improvement in time resolution and angular resolution was achieved at the expense of much greater (apparently by a factor of about 40) random-coincidence rate.

With the discovery[6,7] that the decay curve of positron annihilations in Teflon and other molecular substances could be resolved into three, rather than two, exponentially decaying components, it became more important to obtain a time-angle correlation with improved resolution and statistics. The results of such a measurement should be useful in determining the origin of each component, provided that one could select the angle between the γ rays while simultaneously achieving time resolution and statistics sufficiently good to resolve the three components.

Presumably two of the three components arise from the annihilation of positrons in Ps atoms, and the third comes from annihilation of free positrons, but the identification of each individual component has been in doubt. Tao and Green[8] suggested, on the basis of the effects of various heat treatments of Teflon, that free-positron annihilations contribute to the shortest-lived component (of intensity I_1 and mean lifetime τ_1), and that the previously unnoticed intermediate component (of intensity I_2 and mean lifetime $\tau_2 \sim 1$ nsec) comes from pick-off annihilations of ortho-Ps atoms in "cyrstalline" regions of the sample. On the other hand, Brandt and Spirn[9] measured the temperature dependence of I_1, I_2, and I_3 from 80 to 620 °K, and they found that I_2 is independent of temperature, although I_3, and consequently I_1, have a strong temperature dependence. They concluded that the intermediate component comes from free-positron annihilation, and that ortho-Ps annihilation in crystalline regions contributes to the shortest-lived component. Both groups of investigators agreed that the third component (of intensity I_3 and mean lifetime $\tau_3 \sim 4$ nsec) consists of annihilations of ortho-Ps in amorphous regions of the Teflon.

Measurement of the dependence of I_2 and I_3 on the angle between the annihilation γ rays should provide a direct test of the origin of each component, for the following reason: As mentioned above, the angular correlation of photons from pick-off annihilation of positrons in Ps atoms is broader than the angular correlation resulting from free-positron annihilation. This difference between the angular correlations has been observed[4,5] to cause considerable enhancement of I_3 for "large-angle annihilations – that is, annihilations for which the angle between the γ rays deviated from 180° by a relatively large amount (> 5 mrad). Thus, if I_2 results from free-positron annihilations (Brandt-Spirn hypothesis), it should not be greatly enhanced when large angles are selected, whereas if I_2 results from ortho-Ps annihilation (Tao-Green hypothesis), it should be enhanced as much as I_3 is enhanced at large angles.

In the time-angle correlation measurement reported here the intermediate component is clearly resolved. The results show that the value of I_2 is virtually independent of the angle between the γ rays, whereas the value of I_3 is greatly enhanced, as expected, when "large" angles are selected. This result seems to favor the Brandt-Spirn hypothesis that the intermediate component results from annihilation of free positrons. But there is evidence which casts some doubt on this interpretation, and a third alternative also provides a possible explanation of our results.

EXPERIMENTAL TECHNIQUE

The geometrical arrangement of the apparatus is shown in Fig. 1. Counters 1 and 2 determine the positron lifetime; counters 2 and 3 determine the angle between the annihilation quanta. The positron source, about 5 μC of Na^{22}, is surrounded by the Teflon sample and is placed in a hole in the scintillator of counter 1. This scintillator, a 2-in.-long by 1.5-in.-diam cylinder of Naton 136, detects the 1.28-MeV γ ray which is emitted by the daughter nucleus Ne^{22}, within 10^{-12} sec after the creation of the positron.

Counter 2, which is used with counter 1 to determine the positron lifetime, has a 4-in.-long by 1.5-in.-diam scintillator of Naton 136, while counter 3, which is used only for angular information, has a 6-in.-long by 2-in.-diam scintillator of NaI (Tl) for maximum efficiency.

The selection of angles is done by means of the so-called "long-slit" geometry. Lead blocks containing parallel horizontal slits are placed in front of counters 2 and 3. This arrangement selects the vertical projection of the angle between the γ rays, so that it is sensitive to the vertical component p_z of the momentum of the annihilating pair. (If

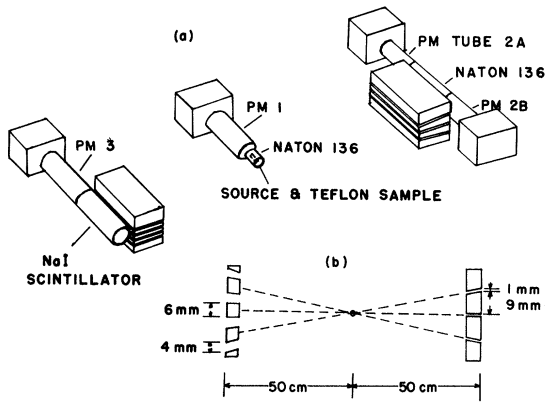

FIG. 1. (a) Geometrical arrangement of counters, slits, and source. Not shown are additional collimating slits near counter 1, and additional shielding around all detectors. (b) Side view of one pair of slits, illustrating angular selection (horizontal scale compressed). In this case, angles θ between 5 and 15 mrad are selected. Dashed lines show possible paths of γ rays for $\theta = 0$.

the vertical projection of the angle between the γ rays is $\pi + \theta$, then $p_z = mc\theta$, to an accuracy of order θ^2.)

Figure 1 illustrates one arrangement of the slits, which selects events for which $\theta \geq 5$ mrad. Events for which $\theta \leq 2$ mrad are selected by replacing the slits shown on counter 3 by another set of slits identical to those shown on counter 2, and aligning the slits so that source and slits are in a straight line. The alignment can be set to a precision of 0.2 mrad simply by counting coincidences between counters 2 and 3 as a function of source height; maximum counting rate indicates alignment of slits and source.

The use of three slits rather than a single slit on each counter increases the counting efficiency with no loss in angular resolution, because the angular separation between one slit and the next is 20 mrad, and the angular correlation of the γ rays (Fig. 2) goes to zero well within 20 mrad of the peak. This arrangement does limit the number of angles for which the slits can be set; in addition to the two ranges of θ mentioned above, one other range was chosen as indicated in Fig. 2.

The long-slit geometry allows one to obtain a large counting rate combined with good angular resolution, but the scintillators used must be much longer than those used in a typical lifetime measurement. Consequently, the time resolution is degraded, because the time required to collect the light from the scintillator depends on the position of the γ-ray interaction. For the 4-in.-long scintillator used in counter 2, the transit time of a light pulse is about 0.5 nsec, a very long time in comparison to the time resolution attainable in the best lifetime measurements [less than 0.3 nsec full width at half-maximum (FWHM) for the "prompt" γ rays of Co^{60}].

In order to overcome this difficulty, we have placed a photomultiplier tube at each end of scintillator No. 2, and we use the time average of the pulses from these two tubes to determine the positron lifetime. This is accomplished by means of a time-to-amplitude converter (TAC) which is essentially two parallel units of the type described by Bell[10] (see Fig. 3). One unit responds to the time difference between pulse from counter No. 1 and from phototube 2*A*; the other does the same for pulses from counter No. 1 and phototube 2*B*. A single capacitor collects the total charge from both units, to produce for a given annihilation a single output pulse whose amplitude is the sum of

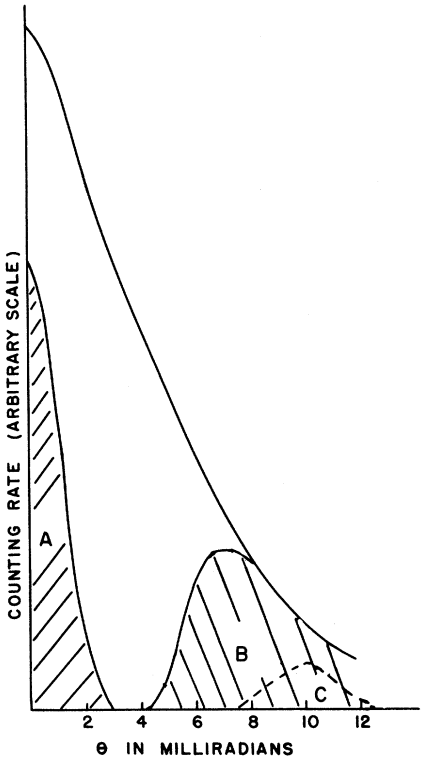

FIG. 2. General shape of angular correlation of γ-ray pairs from positron annihilation in Teflon. Shaded areas indicate the various angular selections used in this experiment; the ordinate of each shaded area is the product of the total height of the curve and the counting efficiency at that angle for a particular pair of slits. Area *A* is defined by a pair of 1-mm slits centered at $\theta = 0$; area *B* is defined by the slits illustrated in Fig. 1; area *C* (enclosed by dashed line) is defined by a pair of 1-mm slits centered on $\theta = 10$ mrad. Note that area *B* contains area *C*.

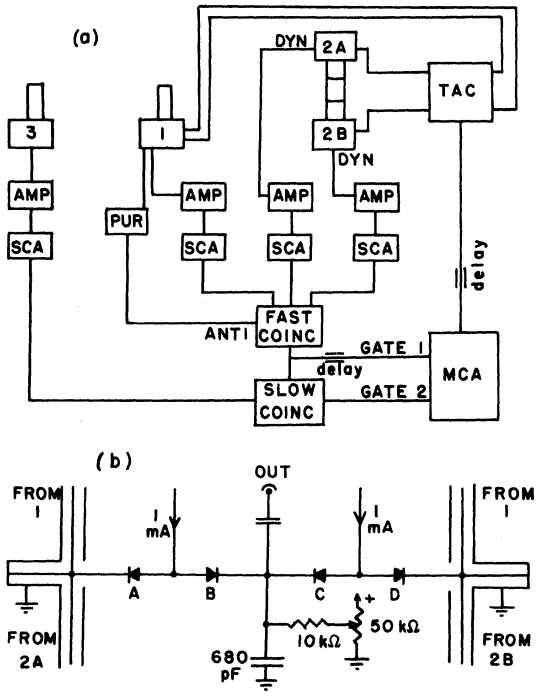

FIG. 3. (a) Block diagram of electronics. TAC receives rectangular pulses from built-in tunnel-diode discriminators on bases of phototubes Nos. 1, 2*A*, and 2*B*. Tube No. 1 has two identical outputs, one of which is used with number 2*A*, the other with number 2*B*. Dynode ouptuts (DYN) on each tube are used to gate the MCA, if pulses are the size required by the single-channel analyzers (SCA). There is also a pileup rejector (PUR) which produces an anticoincidence pulse whenever there are two pulses in counter No. 1 within a 2-μsec interval. Not shown are pulse-height compensation units which are used to "correct" the TAC output for errors resulting from the wide range of pulse heights permitted by the SCA's. (b) Dual TAC circuit. In normal state, constant-current supplies send 1 mA through diodes *A* and *D* to ground. When pulses from 1 and 2*A* are both present, diode *A* is cut off, and the current of 1 mA charges the 680-pF capacitor. When pulses from 1 and 2*B* are both present, diode *D* is cut off, and the other current of 1 mA charges the same capacitor. The output pulse is thus propotional to the time of overlap of pulses 1 and 2*A* plus the time of overlap of pulses 1 and 2*B*.

the pulse amplitudes which would be produced by each unit independently. This summing tends to cancel variations in output pulse amplitude which result from variations in the point of interaction of a γ ray in scintillator No. 2. As a result, a time resolution of 0.8 nsec (FWHM) is achieved, somewhat better than would be attained with a single photomultiplier on a scintillator of only half the size for counter No. 2. In contrast, when only one phototube is used on counter No. 2, the FWHM increases to 1.6 nsec.

Figure 3 also shows a block diagram of the electronics. When there is a coincidence between the single-channel pulse-height analyzers (PHA) on counters 1 and 2, the multichannel analyzer (MCA) is gated to accept the TAC pulse and to store the information in one 200-channel subgroup. When there is a coincidence between the PHAs on all three detectors, the TAC pulse height is recorded in a second 200-channel subgroup. Thus the second subgroup contains a lifetime distribution only for those positrons for which the annihilation quanta are emitted at the selected angle, but one can also find the lifetime distribution of *all* the positrons by adding corresponding channels in the two subgroups. The latter distribution serves as a monitor; this distribution, which involves only counters 1 and 2, should remain unchanged when the slits on counter 3 are changed to select a different angle. After one has verified that this is so, one has confidence that a change in the lifetime distribution of the selected events is truly a result of the difference in the angular selection.

RESULTS

Figure 4 shows the over-all lifetime distribution of positrons in our Teflon sample, as measured without angular selection on a separate apparatus with time resolution of about 0.4 msec (FWHM) for the γ rays from Na^{22}. This curve was used to establish an accurate value for the mean lifetime of each component of the lifetime distribution, so that analysis of the time-angle correlation data could be simplified, as described below.

The values of the mean lifetimes were established by a computer program[11] which constructs a lifetime distribution by assigning a value to the intensity and to the mean lifetime of each component in the distribution, as well as to parameters W and P, where W is the width (FWHM) of the time-resolution function (assumed to be Gaussian) and P is the position of the time $t = 0$ relative to the boundaries between channels of the multichannel analyzer. The constructed distribution is then compared point by point with the observed distribution, and a value of χ^2 is computed for the fit. The parameters can then be varied and new curves constructed and tested until a minimum value for χ^2 is obtained. Errors are assigned by studying the behavior of χ^2 as the parameters are varied in the region near the best fit.

When this program was used to fit the observed lifetime distribution to a three-component distribution, the best values for the intensities were found to be

$$I_1 = 0.675, \quad I_2 = 0.166 \pm 0.005, \quad I_3 = 0.159 \pm 0.002,$$

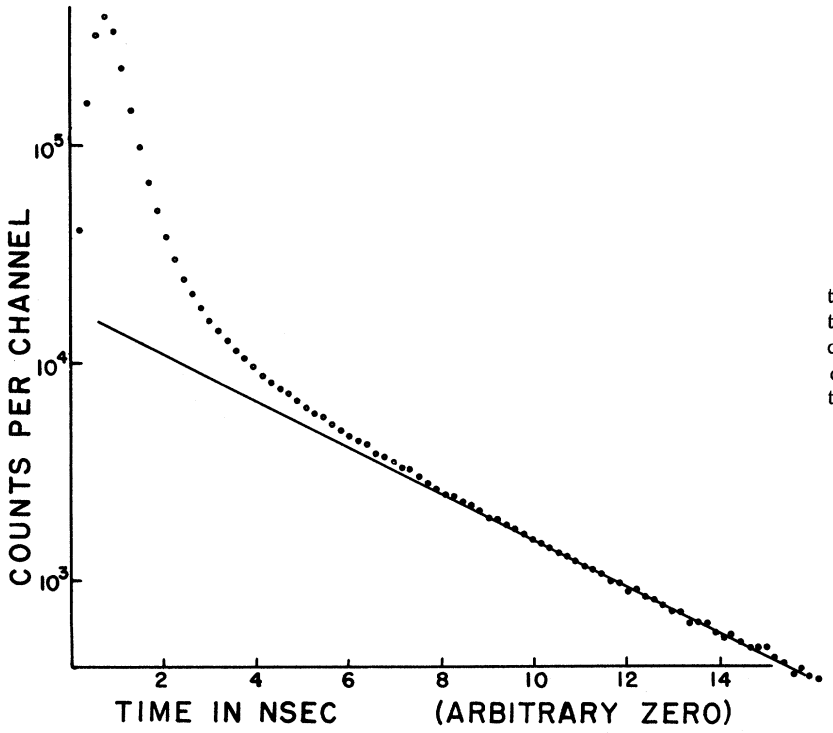

FIG. 4. Over-all lifetime distribution of positrons in Teflon, after subtraction of random coincidences (76 counts per channel). The solid line depicts the slope of the longest-lifetime component.

and the corresponding mean lifetimes were, in nsec,

$\tau_1 = 0.33 \pm 0.01$, $\tau_2 = 1.05 \pm 0.03$, $\tau_3 = 4.06 \pm 0.03$

The errors quoted are statistical only, and do not include possible calibration errors, which would not affect the results to be presented here. The values quoted are in reasonable agreement with other published results on Teflon at room temperature[8,9]; precise agreement is not to be expected, because of the variations in structure from one Teflon sample to another.

The same computer program was used to determine the intensity of each component in the time-angle correlations, except that the computer was instructed to hold the values of τ_1, τ_2, and τ_3 fixed while searching for the best values of I_2, I_3, W, and P to fit the lifetime distribution for each selected angle. This was done to simplify the calculation, because there is no reason to believe that the mean lifetime of a given component should be correlated with the angle between the γ rays; one expects only the intensity to be correlated with angle, because of the difference in angular correlation for the annihilations from the various states. The program was also run with various other fixed values of τ_1, τ_2, and τ_3 so that we could determine how much effect the selection of these parameters had on our final results.

Figure 5 shows the results of a 5-week run (about 43 000 min of counting) during which the angular selection was changed between $\theta \leq 2$ mrad (area A in Fig. 2) and $\theta \geq 5$ mrad (area B in Fig. 2) at intervals of 2 or 3 days. The difference between the two lifetime distributions is quite apparent. The two "monitor" distributions (without angular selection) obtained at the same time are indistinguishable from each other, so it is clear that the difference seen in Fig. 5 is an effect of the change in angle and is not caused by drifts in the electronics over this long time period.

Table I summarizes the results obtained from the analysis of this run ("run 1") as well as those from a 10-week run, ("run 2") in which the narrow slits were used in two different positions – one position centered on $\theta = 0$, as in run 1, to select area A again, and the other position centered on $\theta = 10$ mrad, to select area C. During run 1, 20 000 positron lifetimes were associated with "area A" angles and 19 000 lifetimes were associated with "area B" angles.

During run 2, 15 000 positron lifetimes were associated with "area A" angles and 10 000 lifetimes were associated with "area C" angles.

Table I shows clearly that I_3 is greatly enhanced at large θ, whereas I_2 changes little, if at all, with angle. The ratios of the intensities are

$$\frac{I_2(\text{large-}\theta \text{ events})}{I_2(\text{small-}\theta \text{ events})} < 1.05 \quad ,$$

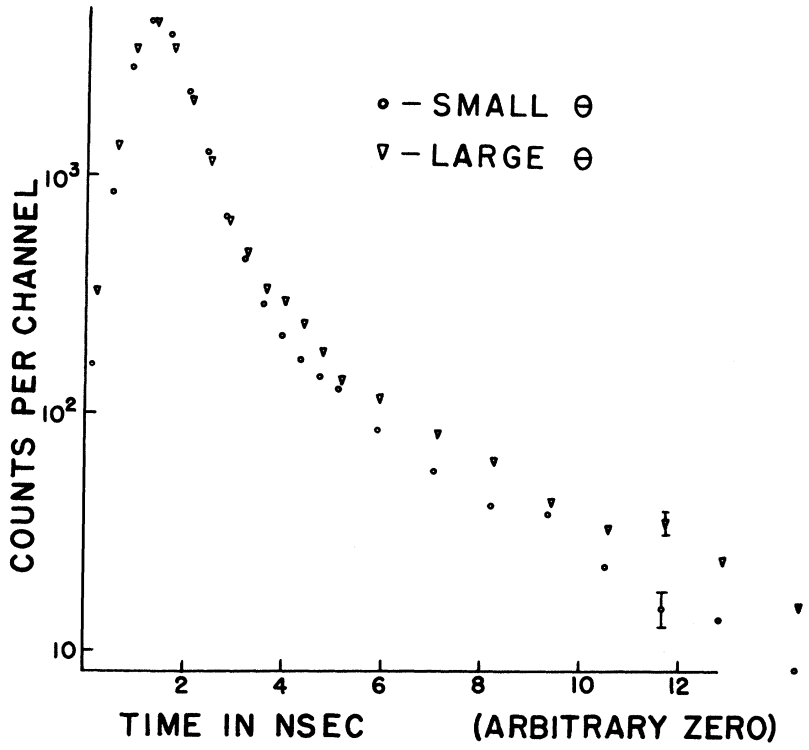

FIG. 5. Distribution of positron lifetimes which were correlated with angle between γ rays, after subtraction of random coincidences (2.7 counts per channel for each curve). Channels containing fewer than 100 counts are averaged in groups of three, for clarity.

$$\frac{I_3(\text{large-}\theta\text{ events})}{I_3(\text{small-}\theta\text{ events})} \sim 1.35$$

The difference between these ratios is well beyond the uncertainties in the measurement, so that the greater angular dependence of I_3 is established. As mentioned in the Introduction, this is a definite indication that I_2 cannot result from ortho-Ps annihilations.

The possibility of a four-component fit to the lifetime distribution was also investigated. If the shortest-lived component results from annihilation of Ps atoms, this component itself must be composed of two or more components, [12] because $\frac{1}{4}$ of the Ps atoms are para-Ps, whose mean lifetime should be equal to or less than the free-space lifetime of 0.125 nsec for para-Ps. This lifetime is quite a bit shorter than the lifetime assigned by the computer program to the shortest-lived component, and the fraction of positrons in this lifetime is considerable. Therefore, it is possible that one might find somewhat different values for the mean lives of the components if one were to fit the over-all lifetime distribution to a four-component curve. A change in the assigned mean lives could possibly affect the computed values of the intensities shown in Table I.

On the basis of the Brandt-Spirn hypothesis, the four components would have intensities as follows: (i) I_4: longest-lived components, resulting from ortho-Ps in amorphous regions; (ii) I_3: second-longest-lived component, resulting from free positrons; (iii) I_2: third-longest-lived component, resulting from ortho-Ps in crystalline regions; and (iv) $I_1 = \frac{1}{4}(1 - I_3)$: shortest-lived component, resulting from para-Ps annihilations.

On the other hand, if the Tao-Green hypothesis were correct, the decomposition of the curve into four components would be as follows: (i) I_4: longest-lived, resulting from ortho-Ps in amorphous regions; (ii) I_3: second-longest-lived, resulting from ortho-Ps in crystalline regions; (iii) I_2: third-longest-lived, resulting from free-positron annihilation; and (iv) $I_1 = \frac{1}{3}(I_3 + I_4)$: shortest-lived, resulting from para-Ps annihilations.

There is still a third alternative. It has been suggested [13] that positrons in Teflon may form bound states with the Teflon molecules. Without

TABLE I. Results of analysis of events into angular areas for two runs.

Run	Mean lifetime (nsec)	Intensity Area A	Area B	Area C
1	1.05	0.245±0.014	0.258±0.014	• • •
	4.06	0.140±0.005	0.185±0.005	• • •
2	1.05	0.252±0.016	• • •	0.255±0.020
	4.06	0.132±0.006	• • •	0.185±0.007

inquiring into the details of this bound state, one might expect that such a state would have a mean lifetime close to the spin-averaged mean lifetime of Ps atoms (0.5 nsec), which is close enough to the observed lifetime to warrant further investigation. According to this hypothesis, the four components would have the intensities: (i) I_4: resulting from ortho-Ps annihilations; (ii) I_3: resulting from positrons bound to Teflon molecules; (iii) I_2: resulting from free positrons; and (iv) $I_1 = \frac{1}{3} I_4$: resulting from para-Ps annihilations.

On any hypothesis, the value of I_1 is determined by the values of I_3 and/or I_4, and the value of I_2 is determined by the condition that $I_1 + I_2 + I_3 + I_4 = 1$, so that there are only two independent intensity parameters, as there are for the three-component fit. Furthermore, one can fix the value of τ_1 to be equal to the para-Ps lifetime in free space, 0.125 nsec, so that no additional adjustable parameter is introduced by the decomposition of the curve into four components rather than three.

Each of the three hypotheses was used to determine a set of mean lifetimes for the over-all lifetime distribution, using the data of Fig. 4 as before. The mean lifetimes thus obtained were then used to determine the intensities of the assumed components in each of the curves of Fig. 5, just as was done for the three-component fit. Although the mean lives and the intensities differed somewhat from the values quoted above for the three-component fits, the general result was unchanged: The intensity of the longest-lived component was enhanced by about 30% at larger θ, whereas the intensity of the next-longest-lived component, the so-called intermediate component, was enhanced by less than 5%, if at all, at large θ relative to small θ.

One interesting feature appeared when the over-all lifetime distribution was decomposed into four components; it was found that the various hypotheses were not equally able to fit the over-all distribution. The best values of χ^2 obtained for the different methods of decomposing the curve are shown in Table II.

DISCUSSION AND CONCLUSIONS

The Tao-Green hypothesis is rather decisively ruled out by the time-angle correlation results. But two features of the data are somewhat puzzling.

First, the different values of I_2 shown in Table I are *all* greater than the value of $I_2 = 0.166$ quoted for the over-all lifetime distribution. This could be the result of some systematic error in the analysis of the lifetime distributions. The choice of resolution function is the most logical candidate for the origin of such a systematic error. The Gaussian resolution function, which fits a "prompt" curve quite well, is probably not appropriate for a 5-week-long run. For such a long run, drifts in the position of the peak channel make a sizable contribution to the width of the curve.

An attempt was made to compensate for such drifts by using a resolution function whose central part was flatter than a Gaussian. When this resolution function was used, the results indicated that it was a better choice; the values of I_2 which resulted were smaller than those of Table I, although they were still not so small as 0.166. But the final result was unchanged; the angular dependence of I_2 was still found to be almost nil.

Second, it is somewhat surprising that the Tao-Green hypothesis yields the smallest value of χ^2 for the four-component fit to the over-all lifetime distribution. However, the values of χ^2 shown in Table II cannot be interpreted literally as an indication of the relative correctness of the various models, because there are a number of factors which could cause the observed data to deviate from the "ideal" computer-constructed curves. These factors include: (i) differential nonlinearity in the TAC, which was less than 2%, but which could easily have an effect on the decomposition of the curve into four components; (ii) changes in the annihilation rate during the thermalization time of the positrons, which would change the shape of the curve over a time of order 10^{-10} sec; (iii) possibility of scattering from one counter to another, which would produce additional prompt counts in the lifetime distribution; and (iv) the necessity of digitizing the time resolution in constructing the curves in the computer program, which could have a subtle effect on the shape of the curve in the region very near to time $t = 0$.

These effects are all quite small, but they could have a significant effect on the value of χ^2 when one is dealing with a curve containing 400 000 counts in the peak channel. For example, differential nonlinearity of 2% could change the peak counting rate by about 12 standard deviations under

TABLE II. Comparison of hypotheses.

Method of fitting the over-all lifetime distribution	Assumed origin of intermediate component	Best value of χ^2 (90 deg of freedom)
Three-component fit		160
Four-component fit, $I_I = \frac{1}{4}(1 - I_3)$	Free positrons	488
Four-component fit, $I_1 = \frac{1}{3}(I_3 + I_4)$	Ortho-Ps	113
Four-component fit, $I_1 = \frac{1}{3} I_4$	Bound state with Teflon	143

these conditions. The thermalization time of the positrons is not known, and even if it were, one does not know what sort of reactions might occur during that time which would affect the annihilation rate. Finally, scattering of γ's from one counter to another was minimized by the use of shielding and placement of counters, but it could not be entirely eliminated; a certain amount of scattering is inevitable when one places the counters close enough to each other to obtain a large counting rate.

Thus one must be prepared to accept a rather large value of χ^2 in fitting the data, and one can attach no special significance to the small value shown for the Tao-Green hypothesis. One simply cannot decide among the various hypotheses on the basis of the lifetime distribution alone.

However, the value of $\chi^2 = 488$ found for the Brandt-Spirn hypothesis does seem to be a bit excessive, even after one allows for contributions from the above sources. For this reason we would tend to support the hypothesis of a bound state between the Teflon molecule and the positron, which provides a sufficiently good fit to all the data, and we suggest that further work be done with this hypothesis in mind.

ACKNOWLEDGMENTS

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[11]Developed by R. Eaton and J. McGervey.

[12]Strictly speaking, there are *five* lifetime components, because the pickoff annihilation rates, and hence the para-Ps lifetimes, are different in the two regions of the Teflon. But there is little chance that these two shortest lifetimes could be distinguished from each other with the time resolution quoted here.

[13]J. W. Weinberg (private communication).