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On the Nature of the Medium Lifetime Component of Positron Annihilation in a Teflon Polymer

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The nature of the medium lifetime component of positron annihilation in a Teflon (PTFE) polymer has been investigated by using a recently developed Time Selecting γ Energy Spectrometer with and without a magnetic field. It has been assigned to o-Ps quenching annihilation, just the same as the longest lifetime component.

1.INTRODUCTION

In a molecular substances, especially in organic polymers, positron annihilation lifetime spectrum is rather complex because of the formation of the e^+e^- bound state, i.e. the positronium atom. Within the present time resolution ~ 200 ps of the lifetime spectrometer, the lifetime spectrum can generally be decomposed into 4 lifetime components with the least-squares fitting method (see Table 1 and Ref.[1]). In the shortest component τ_1(100 ps), except for the contribution of the self-annihilation of the singlet positronium (p-Ps), there could be another contribution whose origin is not yet known; the second one τ_2(350 ps) has been ascribed to positron free annihilation with a relative intensity up to 50%; the longest component τ_4 has been assigned to the pick-off quenching of the triplet positronium (o-Ps) with a value of 2--4 ns at room temperature depending on the composition and structure of the polymers. As to the medium lifetime component τ_3 which has a

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value of 1.0--1.3 ns for the flexible chain polymers and some small molecule matter, there have been such speculations and arguments about its annihilation nature and mechanism since the 1960's[2] such as positron free annihilation[3--5], positron trapping in the defects of the media[6,7], positron attaching onto the molecule[8--10], and so on. Recent results[11--13], including 3γ coincidence measurement by Dauwe[11] tend to ascribe it to o-Ps quenching which is the same as the early speculation[14]. However, all these arguments are indirect speculations.

Since the 1980's, Zhang Tianbao[15,16] has developed a Time Selecting γ Energy Spectrometer (abbreviated to TSES) and improved the BaF_2 detector lifetime spectrometer[17]. We have used the new spectrometers, especially the TSES with and without an applied magnetic field to perform a measurement from which direct evidence for the nature of the medium lifetime component may be obtained. Clarification of this matter will help us further to understand positron behavior in polymers and other molecular materials, and investigate the physical and chemical properties of the media by the positron annihilation technique.

2. EXPERIMENTAL PRINCIPLE AND METHOD

In antimagnetic matter, o-Ps atoms (3/4 of the whole positronium atoms) interact with the electron cloud of the molecules and undergo pick-off quenching with a rate of $\lambda_n > \lambda_t$, where λ_t is the self annihilation rate of o-Ps in vacuum:

$$\lambda_t = \frac{1}{\tau_t} \approx \frac{1}{140}\ (\text{ns})^{-1}$$

$$\lambda_s = \frac{1}{\tau_s} \approx \frac{1}{0.125}\ (\text{ns})^{-1},$$

Under an applied magnetic field with strength H, the $m = 0$ magnetic substate of o-Ps (1/3 of o-Ps atoms) will suffer a magnetic perturbance by mixing with the $m = 0$ state of p-Ps and be terminated with 2γ decay, which is called magnetic quenching. The total annihilation rate will be λ' rather than the original value λ in the $H = 0$ case. The $m = \pm 1$ substate of o-Ps does not suffer the magnetic perturbance. Its total rate λ is independent of the magnetic field. We have the following expressions:

$$\lambda = \lambda_n + \lambda_t \approx \lambda_n \qquad H = 0,\ m = 0, \pm 1; \tag{1}$$

$$\lambda = \lambda_n + \lambda_t \approx \lambda_n \qquad H \neq 0,\ m = \pm 1; \tag{2}$$

$$\lambda' = \frac{y^2(\lambda_s + \lambda_n) + \lambda}{1 + y^2} \approx \frac{y^2(\lambda_s + \lambda_n) + \lambda_n}{1 + y^2} \qquad H \neq 0,\ m = 0, \tag{3}$$

where $y = [(1 + x^2)^{\frac{1}{2}} - 1]/x$, $x = 4\mu H/\Delta W = H/36.5$ with μ standing for the Bohr magneton, ΔW the hyperfine splitting energy between o-Ps and p-Ps ground states, H being the field strength in kilo-Gauss and $\lambda_s = 1/\tau_s = 1/0.125\ \text{ns}^{-1}$ referring to the self annihilation rate of p-Ps.

According to Eqs.(1) to (3), it can be seen that a new lifetime component $\tau' = 1/\lambda'$ appears under $H = 0$ which makes the lifetime spectrum more complicated. If there are more than one o-Ps

TABLE 1. Positron Annihilation Lifetime Components in Teflon

H (kG)	FWHM (ps)	τ_1	τ_2	τ_3	τ_4	I_1	I_2	I_3	I_4	Program *
		(ns)				(%)				
0	200.0	0.118	0.352	1.395	4.252	18.5	48.4	14.3	19.2	EXP(S)
0	200.0	0.117	0.355	1.408	4.272	22.4	47.7	12.8	17.3	EXP(G)
0	195.50	0.114	0.352	1.449	4.292	16.1	50.4	14.6	18.8	POS(S)
0	650.0	—	0.357	1.315	4.171	—	73.2	11.9	14.9	EXP(G)
18.5	650.0	—	0.352	1.163	3.815	—	77.7	13.0	9.3	EXP(G)

*EXP: Multi-exponential fitting method[19], Program EXPFIT (unpublished)
POS: time resolution convolution fitting program[20]
S: analysis results of single lifetime spectrum
G: average of analysis results of lifetime spectra

lifetime components (such as in the polymer media) which undergo the magnetic quenching, the analysis of the lifetime spectrum would be impossible. On the other hand, good information can be extracted from the analysis of the Doppler broadening energy spectrum of positron 2γ annihilation. A part of λ', that is,

$$P = \frac{y^2(\lambda_s + \lambda_n)}{(1 + y^2)\lambda'} \qquad (4)$$

will contribute a narrower component in the Doppler broadening spectrum due to the smaller momentum of Ps atoms. For example, if p-Ps as well as o-Ps is fully thermalized before its annihilation, the width of the narrow peak which is from p-Ps decay or o-Ps magnetic quenching can be decreased to FWHM ≤ 1 mrad in 1D-ACAR curve at room temperature. The remaining part of λ', $P_r = 1 - P \approx \lambda_n/(1 + y^2)\lambda'$, will contribute a broad component (FWHM ~ 10 mrad in 1D-ACAR curve), because this part originates from o-Ps pick-off quenching in which the positron in o-Ps eventually annihilates with an energetic electron bounded in a medium molecule. For a Ge detector with the energy resolution FWHM = 1--1.5 keV (equivalent to an angular resolution ~ 4 mrad for 1D-ACAR), the difference between the two above Doppler components is also distinct. The TSES with the magnetic quenching method is effective for studying the correlation between the lifetime spectrum and the narrow component of the Doppler spectrum.

TABLE 2. Some Theoretically Calculated Values

H(kG)	15	18	20	36.5
$\tau_3' = 1/\lambda_3'$(ns)	0.94	0.85	0.80	0.52
$\tau_4' = 1/\lambda_4'$(ns)	1.85	1.53	1.37	0.71
P_3 (%)	31	38	43	66
P_4 (%)	57	65	69	86

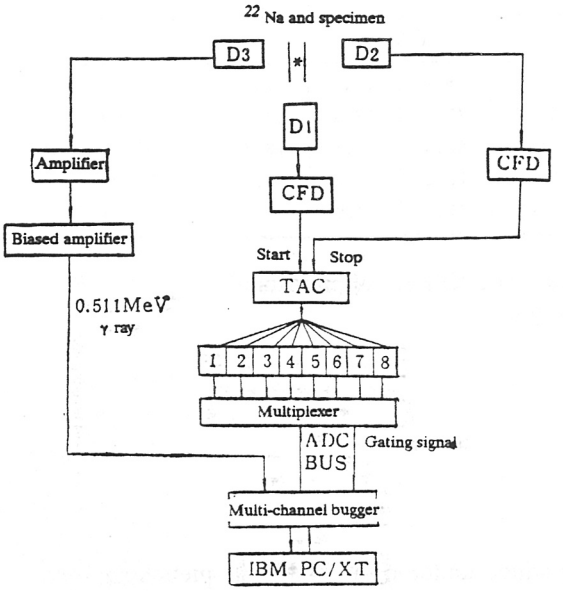

FIG. 1
Diagram of time-selecting energy spectrometer.

D_1: plastic scintillator detector (ST-401, 30×40×70 mm^3)
D_2: BaF_2 detector ('50 mm × 40 mm)
D_3: HPGe detector
CFD: Constant Fraction Discriminator
1--8: SCA

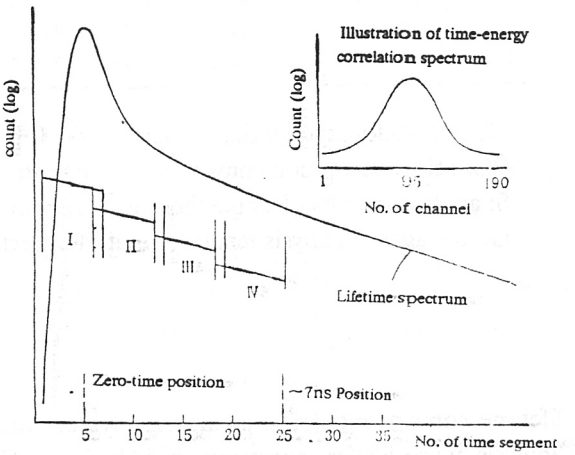

FIG. 2
Illustration of experimental method.

I-IV stand for the four time ranges.

Supposing that the medium component τ_3 and the longest one τ_4 (under 4 component fitting) of positron annihilation in the teflon polymer are both of o-Ps quenching, the magnetic quenching components $\tau_3' = 1/\lambda'_3$ and $\tau'_4 = 1/\lambda'_4$ should appear in the lifetime spectrum, which are correlative with the narrow components P_3 and P_4 in the Doppler spectrum. From Eqs.(3) and (4) the values of $\tau_3 = 1/\lambda_{n3}$ and $\tau_4 = 1/\lambda_{n4}$ listed in Table 1 (a 3 component fitting result is listed in the 4th and 5th rows for comparison), theoretical values of τ'_3, τ'_4, P_3 and P_4 are evaluated and presented in Table 2. It can be seen that the higher the H, the larger the P_3 and P_4, and so the easier to extract the narrow Doppler component. However, as the values of τ'_3 and τ'_4 become closer, it will be difficult to separate them. Therefore $H \sim 20$ kG seems to be a good compatibility.

The TSES equipment is shown in Fig.1. It consists of a high purity germanium detector (HPGe, D_3) combined with a lifetime system (D_1 and D_2). HPGe is used to measure the Doppler broadening of the 0.511 MeV annihilation γ and its energy resolution is 1.02 keV for ^{85}Sr 0.514 MeV γ ray. Through 8 SCA's (Single Channel Analyzer) and a Multiplexer/Router, the lifetime system

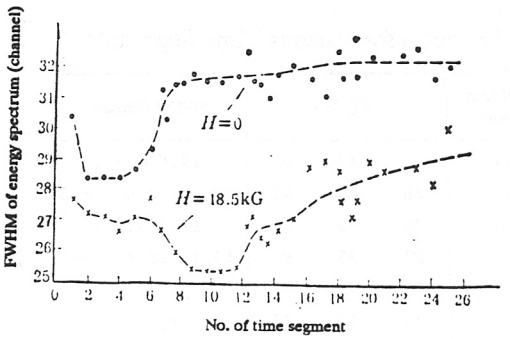

FIG. 3
Apparent FWHM of time-energy correlation spectrum.

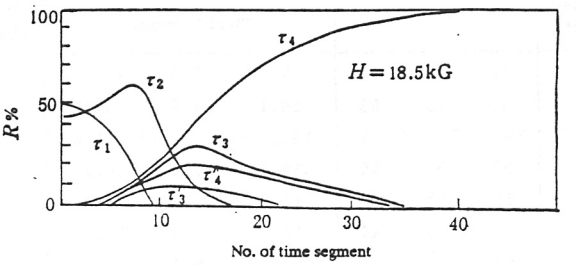

FIG. 4
Simulating results of $R\%$.

allows simultaneous observation of 8 time windows corresponding to the different ages of o-Ps. 8 Doppler-broadened spectra correlated with different ages of o-Ps are simultaneously measured in 8 segments of a 8192 multichannel buffer. H = 18.5 kG was used for the magnetic quenching measurement, and the time resolution of the system is 0.8 ns in this condition. The triple coincidence counting rate is ~ 50 cps for a ^{22}Na source (5--8 μci).

3.MEASUREMENT AND DATA TREATMENT

The main procedure of the measurement for the time selecting Doppler spectra of positron annihilation in teflon is shown in Fig.2. The chosen time range was from ~1.5 ns before zero time (the peak position of the lifetime spectrum) to ~ 7 ns after. The measurement was done in 4 runs. The time region selected in the 1st run was from ~ 1.5 ns to 0.9 ns and 0.5--2.9 ns, 2.5--4.9 ns and 4.5--6.9 ns for the 2nd, 3rd and 4th runs respectively. Each region (2.4 ns) was divided into 8 time intervals (0.3 ns each) which correlated to 8 Doppler spectra counted in different segments of multichannel buffer. Measurement, in each run contained two groups of Doppler spectra accumulated separately with and without a magnetic field.

Fig.3 shows the average apparent FWHM (no deconvolution for the response function of the spectrometer) of all the time selected Doppler spectra. The dash lines represent the trends of FWHM with the number of time segments. It can be seen that the effect of FWHM becoming narrower under H = 18.5 kG is very obvious. Furthermore, the time region corresponding to the narrower FWHM's is about the same as that expected by a simulating calculation as shown in Fig.4. The longitudinal coordinate $R\%$ in Fig.4 stands for the intensity proportion of every lifetime component in each time segment (under H = 18 kG). τ_3 and τ_4 were supposed to be of o-Ps quenching origin, so the parameters listed in Tables 1 and 2 were used in our calculation.

To make an exact judgement of the annihilation nature of τ_3 in teflon, it is necessary to find the amount S (such as the total counts) arising from the magnetic quenching of o-Ps in each time

TABLE 3. F values and FWHMs of Differential Spectra for Various Time Segments

No. of time segment*	F(%)				FWHM Channel				No. of time segment*	F(%)			FWHM Channel
1	80	83	86		17.6	17.6	15.7		10	86	89	92	19.5—13.2
2	80	83	86		16.1	16.4	16.4		11	86	89	92	13.8 18.6 16.3
3	80	83	85	87	16.3	16.0	18.5	16.8	12	86	89	92	17.8—13.6
4	80	83	86		16.2	15.6	14.3		13	92	95	98	18.4 15.4 —
5	80	83	86		16.6	16.3	14.2		14	92	95	98	<19**
6	80	83	86		17.1	15.6	17.2		15	92	95	98	<19
7	83	86	89		18.0	16.2	12.4		16	92	95	98	<19
8	86	89	92		15.0	15.0	12.5		17	92	95	98	<19
9	85	87	90		16.8	17.2	15.1		18	92	95	98	<19

*Point No. agrees with the datum point in Fig.6.

**The fitting value is not shown due to the low counting statistics, but the upper limit is known.

segment and to draw a curve of log S vs. No. of time segment to see if it fits the law of exponential decay. According to the primary analysis shown in Fig.3 and a deconvolution Gaussian fitting[21] for the measured spectra, we think that the Doppler spectrum measured with magnetic a field is approximately a superposition of the spectrum (S = counts of the spectrum) originating from the

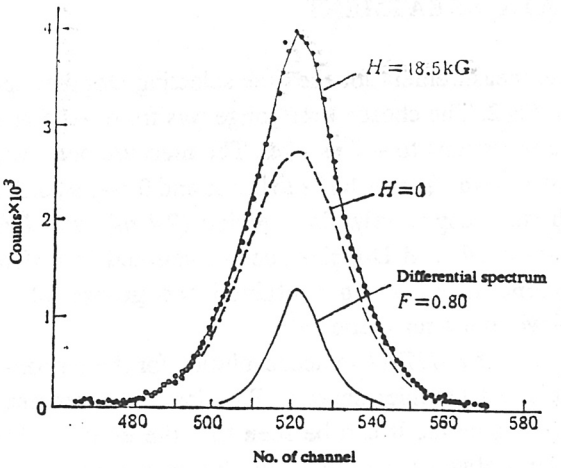

FIG. 5

An example of obtaining the amount of the narrower component energy spectrum by subtraction.

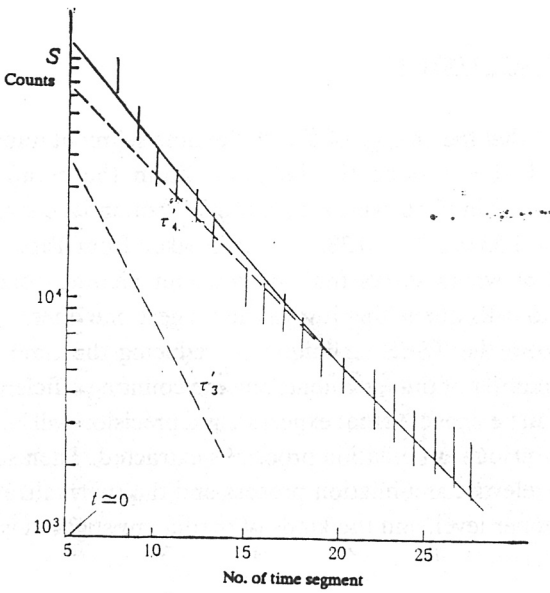

FIG. 7
Plot of log S vs.No. of time segment.

o-Ps magnetic quenching and the spectrum measured without magnetic field, constricted to a certain proportion A which is of course different from segment to segment. The remaining part A-1.00 is called F factor here. A reasonable value of S must be dependent on the selection of the F value. In view of the complexity of the lifetime spectrum of teflon and the poor counting statistics in the important time region (around 2 ns), the reasonable S value for each time segment has been extracted by means of a practical procedure, which is: the Doppler spectrum without magnetic quenching is normalized by the total counts of the spectrum with magnetic field measured in the same time segment; the normalized spectrum multiplied by an F factor, which is then subtracted from the spectrum measured under the magnetic field to produce a differential spectrum; by adjusting F and finding out an interval of F value where the differential spectrum can be fitted to a single Gaussian, the Gaussian FWHM is calculated and finally the Gaussian area counted to get S value. The above procedure is repeated for all the time segments. For example, one of the differential spectrum with F = 80% is shown in Fig.5. All of the F values and Gaussian FWHM values are listed in Table 3 and the final experimental result log S vs. No. of the time segment is shown in Fig.6. The relative experimental error of S value is estimated to be ~ 30% including the deviations of counting statistics, the electronic shift of the TSES, the fluctuation of magnetic field, the approximation of various hypotheses and some man-made regulations in data processing. We tried to fit each of the differential spectra with double Gaussian and found that for a small value of F, the width (~ 40 channel on segment) of the wider Gaussian is close to the width of the spectrum without magnetic

quenching and the width (~ 16 channels) of the narrower Gaussian is close to the FWHM values in Table 3. But the double Gaussian fittings are always divergent in the chosen intervals of F value. Besides, as expected, the chosen F presented in Table 3 increases slowly with the increase of the segment number.

4. DISCUSSION AND CONCLUSION

It can be seen from Fig.6 that the change of S with the time segment number (corresponding to the decay time) shows a double exponential behavior. From the comparison between the experiment data and the solid curve in Fig.6, which is a double exponential curve calculated with the parameters τ'_3 = 0.85 ns, τ'_4 = 1.53 ns, P_3 = 0.38, P_4 = 0.65 taken from Table 2 (teflon at H = 18 kG), there is a good agreement which shows that the medium lifetime component of positron annihilation in teflon belongs to o-Ps quenching just as the longest one does.

It is necessary to improve the TSES technique by reducing the time resolution with the magnetic field, increasing the number of time segments and the counting efficiency, and reducing the random background. Thus, it can be expected that experimental precision will be improved and more fine energy information about various annihilation processes extracted. Then some basic problems such as the mechanism of the relevant annihilation process and the thermalization dynamics of Ps atoms can be explored on a deeper level, and the kinds of media investigated extended.

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