Static Magnetic Field Quenching of the Orthopositronium Decay: Angular Distribution Effect*

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An effect of the angular dependence of the annihilation radiation from the magnetic substates of orthopositronium has been observed in an experiment similar to that of Deutsch and Dulit. Quanta due to positron annihilations were observed at a direction of 90° to a magnetic field. The fraction, f, of orthopositronium decaying by three-quantum annihilation can then be computed, and at high values of the field f was found to be less than $\frac{2}{3}$. The value $\frac{2}{3}$ is expected statistically since only annihilation radiation of the $M=0$ substate is affected by the field. Our observation is understandable if Drisko's formulas for the angular distribution of the annihilation radiation from the different magnetic substates are considered.

The ratio of the decay rates of parapositronium to orthopositronium can be computed from values of f *vs* the field, after corrections are made for the angular distribution of the quanta. Our data yield $\lambda_p/\lambda_o=1302$ with a probable error of ±15 percent, in agreement with the theoretical value of 1114.

1. INTRODUCTION

THE mode of annihilation of ground state ($n=1$) positronium is known to be affected by a static magnetic field. In the absence of a magnetic field positronium exists either in the para-state ($^1S_0$, $M=0$) or in the ortho-state ($^3S_1$, $M=0, \pm1$). From the conservation laws of energy, linear momentum, angular momentum, and a component of angular momentum, it follows that a parapositronium atom at rest will decay by the emission of two quanta which have the same energy (mc^2), which propagate in opposite directions, and which are polarized in mutually perpendicular directions.[1,2] An orthopositronium atom at rest will decay by the emission of three quanta whose energies sum to $2mc^2$; the energy distribution of the annihilation quanta has been calculated from conventional quantum electrodynamics. The three quanta will propagate in a single plane, and the directions of propagation and polarization will be such as to conserve linear and angular momentum and a component of angular momentum. The decay rate of parapositronium (λ_p) is 8.03×10^9 sec^{-1} and of orthopositronium (λ_o) is 7.21×10^6 sec^{-1}.[3]

In the presence of a magnetic field the $M=\pm1$ magnetic substates of orthopositronium are still pure ortho-states, and will decay by the three quantum annihilation characteristic of orthopositronium decay. On the other hand, the $M=0$ state of orthopositronium has a small admixture of para-state due to the interaction with the magnetic field, and hence can decay either by three-quantum annihilation or by two-quantum annihilation. The relative probabilities of these two modes of decay depends, of course, on the amount of admixture of para-state and on the ratio of the decay rates of parapositronium to orthopositronium. The $M=0$ state of parapositronium will similarly have a small admixture of ortho-state, but since the orthopositronium decay rate is so small compared to the parapositronium decay rate, this $M=0$ state will still decay predominantly by two-quantum annihilation for practical values of the magnetic field. Thus the principal effect of a static magnetic field is to increase the amount of two-quantum annihilation relative to three-quantum annihilation. In a known magnetic field the amount of quenching of three-quantum annihilation will depend on two parameters: (1) the energy difference between parapositronium and orthopositronium, (2) the ratio of the decay rates of parapositronium to orthopositronium.

The original experiment of Deutsch and Dulit[4] on the static magnetic field quenching of orthopositronium decay was done by a study of the energy distribution of the γ-ray annihilation radiation as a function of the magnetic field. Ore and Powell's theoretical value of the ratio of the decay rates of parapositronium to orthopositronium was assumed, and the experimental data were then used to derive a value for the energy difference between parapositronium and orthopositronium to an accuracy of about 10 percent. Pond and Dicke[5] studied the magnetic quenching of orthopositronium decay by observing the number of two-quantum annihilations by coincidence counting. If one assumes the theoretical ratio of parapositronium to orthopositronium decay rates, their data yield the energy difference to an accuracy of about 25 percent. More recently Deutsch, Brown, and Weinstein[6] have measured the energy separation between parapositronium and orthopositronium by inducing a radio-frequency transition between the magnetic sublevels

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[6] M. Deutsch and S. C. Brown, Phys. Rev. **85**, 1047 (1952); Weinstein, Deutsch, and Brown, Phys. Rev. **94**, 758 (1954); **98**, 223 (1955).

of orthopositronium, and report a value $(2.03370 \pm 0.00050) \times 10^5$ Mc/sec. Wheatley and Halliday[7] measured the quenching of three-quantum annihilation by observing the change in the triple coincidence rate with magnetic field. Using the value obtained for the energy separation from the radio-frequency experiment, they give a value for the ratio of parapositronium to orthopositronium decay rates of 1050 ± 140, compared with Ore and Powell's theoretical result of 1114.

The present paper reports the effect of the angular dependence of the annihilation radiation of the magnetic substates of orthopositronium on the observed magnetic quenching of orthopositronium in an experiment similar to that of Deutsch and Dulit. Recently Drisko has redone the calculation of Ore and Powell on the annihilation radiation from orthopositronium, but without averaging over angles and has thus obtained angular distribution formulas.[8] Our experimental data are in good agreement with his calculation. An effect of the angular dependence of the annihilation radiation from the magnetic substates of orthopositronium in agreement with Drisko's calculation was also observed by Wheatley and Halliday.[7]

If the value of the energy separation is taken from the recent radio-frequency experiment, the present experiment gives the ratio of parapositronium to orthopositronium decay rates. The angular distribution effect must be considered if a value for this ratio is to be obtained which is correct even as to sign and order of magnitude. The value obtained is 1302 with a probable error of ± 15 percent, which is in adequate agreement with the theoretical value of 1114. A preliminary report on this work has been given.[9]

2. THEORY OF THE EXPERIMENT

The part of the Hamiltonian for positronium which includes the "hyperfine structure" interaction (which leads to the energy difference between parapositronium and orthopositronium) and the interaction with the magnetic field is

$$\mathcal{H}=\mathcal{H}_0+g^-\mu_0\mathbf{s}^-\cdot\mathbf{H}+g^+\mu_0\mathbf{s}^+\cdot\mathbf{H}, \tag{1}$$

in which $\mathcal{H}_0$ is the spin-dependent "hyperfine structure" operator,[10] s^- and s^+ are the spin angular momentum operators for the electron and positron respectively, $g^-(\simeq 2)$ and g^+ are their spin gyromagnetic ratios, μ_0 is the Bohr magneton, and $H=H_z$ is a constant magnetic field applied in the z direction. Note further that $g^+=-g^-$. To first-order perturbation theory in the magnetic interaction the state functions for a field H, which correspond adiabatically to the zero field states $J=1$, $M=0$ and $J=0$, $M=0$, are, respectively,

$$\begin{aligned}\psi_1&=u(1,0)+au(0,0),\\ \psi_0&=u(0,0)-au(1,0),\end{aligned} \tag{2}$$

in which J is the total angular momentum quantum number, M is the quantum number for component of total spin angular momentum in the z direction, $u(1,0)$ and $u(0,0)$ are the zero-field eigenstates for $J=1$, $M=0$ and for $J=0$, $M=0$, respectively. The quantity $a=\mu_0 g^- H/W$, in which W—a positive quantity—is the energy difference between orthopositronium and parapositronium.

Halpern[11] has given a discussion of the theory of the quenching of the orthopositronium decay in a constant magnetic field. Upon using his results the fraction, F, of $M=0$ orthopositronium atoms which will decay by three-quantum annihilation in a magnetic field is

$$F=\frac{(1-a^2)\lambda_o}{(1-a^2)\lambda_o+\lambda' a^2}, \tag{3}$$

in which $\lambda'=\lambda_p-\lambda_o\simeq\lambda_p$. The two-quantum decay of $M=0$ parapositronium is negligibly affected by the presence of the magnetic field, as was pointed out in the introduction. The experimental data can be analyzed to yield the fraction, f, of orthopositronium atoms which will decay by three-quantum annihilation. The quantity f is given by

$$f=\frac{2}{3}+\frac{F}{3}=\left(1+\frac{\lambda_p}{\lambda_o}\frac{2}{3}\frac{a^2}{(1-a^2)}\right)\Big/\left(1+\frac{\lambda_p}{\lambda_o}\frac{a^2}{(1-a^2)}\right). \tag{3'}$$

For high fields f approaches the value $\frac{2}{3}$, which is expected statistically since only the $M=0$ ortho-state is affected by the magnetic field.

The angular distributions of the annihilation quanta from the three-quantum decay of the different magnetic substates of orthopositronium are required for the interpretation of the experimental data. Drisko has repeated the calculation of the three-quantum annihilation of orthopositronium without integrating over angles or averaging over the three spin states, and has thus developed formulas which give the relative number of quanta per unit solid angle and per unit frequency interval for the single quanta emitted by the magnetic sublevels of orthopositronium. His formulas are[8]

$$\begin{aligned}&(M=0):\ \tfrac{1}{2}[\sin^2\theta\Psi(k)+\tfrac{1}{2}(3\cos^2\theta-1)\Phi(k)],\\ &(M=+1 \text{ or } M=-1):\\ &\qquad\tfrac{1}{4}[(1+\cos^2\theta)\Psi(k)-\tfrac{1}{2}(3\cos^2\theta-1)\Phi(k)],\end{aligned} \tag{4}$$

in which θ is the angle between the quantization axis (direction of $\mathbf{H}$) and the direction of observation of the quantum, and k is the energy of the quantum in

[7] J. Wheatley and D. Halliday, Phys. Rev. **88**, 424 (1952).
[8] We are grateful to Professor G. C. Wick and Dr. R. Drisko for prepublication information on this calculation. R. Drisko, Phys. Rev. **95**, 611(A) (1954), and article to be published in Phys. Rev.
[9] Marder, Hughes, and Wu, Phys. Rev. **95**, 611(A) (1954).
[10] J. Pirenne, Arch. sci. phys. et nat. **29**, 265 (1947).
[11] O. Halpern, Phys. Rev. **94**, 904 (1954).

units of mc^2. The functions $\Psi(k)$ and $\Phi(k)$ are

$$\Psi(k)=8\left[\frac{2-k}{k}+\frac{2(1-k)}{k^2}\ln(1-k)+\frac{(1-k)k}{(2-k)^2}-2\frac{(1-k)^2}{(2-k)^3}\ln(1-k)\right],$$
$$\Phi(k)=8\left[\frac{k}{2(2-k)^2}-\frac{(1-k)}{(2-k)^3}\ln(1-k)-\frac{4(1-k)}{k^3}-\frac{(2-k)^2}{2k^3}-\frac{3(2-k)(1-k)}{k^4}\ln(1-k)\right]. \quad (5)$$

$\Psi(k)$ is the energy spectrum given by Ore and Powell.[3] The angular distribution of the quanta is independent of ϕ, the azimuthal angle measured about the z-axis.

3. EXPERIMENTAL ARRANGEMENT

The principal features of the experiment are similar to those of Deutsch and Dulit.[4] In the present experiment the positron source was Cu^{64} produced by neutron bombardment of 99.9 percent pure copper in the Brookhaven pile. The source was never used sooner than 8 hours after its production because of the time for delivery and the time for setting up the experiment; this necessary delay, of course, eliminated any possible short-lived impurity radiations. The source strength at the beginning of a run was customarily about 50 mC. The form of the source was a disk $\frac{1}{2}$ in. in diameter and 0.001 in. in thickness.

The experimental arrangement is shown in Fig. 1. The electromagnet has a 9-in. pole face and a gap width of about 3 in.; it is powered from eight 3000-ampere-hour 2-volt submarine batteries, and is capable of producing fields up to about 7000 gauss. Rose shims were placed on the pole faces, and nuclear resonance absorption measurements indicated that the field was uniform to within about 1 percent throughout a cylindrical volume between the pole faces which had a radius of 3 in. measured from the axis of the magnet. A circular brass cylinder 6 in. in diameter and $2\frac{1}{2}$ in. in height is supported between the pole faces with its axis along the magnet axis. The positron source is glued with Duco cement to the center of one of the parallel ends. A substantial amount of lead shielding is included within the cavity so that there are no direct paths from the source to the portion of the side wall that is viewed by the scintillation spectrometer. In this way the amount of annihilation radiation, which reaches the detector because of positrons annihilating in the brass walls, is minimized. The cavity is connected to a pumping system consisting of a VMF 50 oil diffusion pump and a mechanical pump. Because of an unfortunate history, rather long thin tubing is involved so that the pumping speed for evacuating the cavity is only about 3 liters/sec. Customarily the cavity is evacuated to a pressure of about 10^{-3} mm before being filled with the desired gas. Gas pressures in the cavity of between 1 and 2 atmospheres are used.

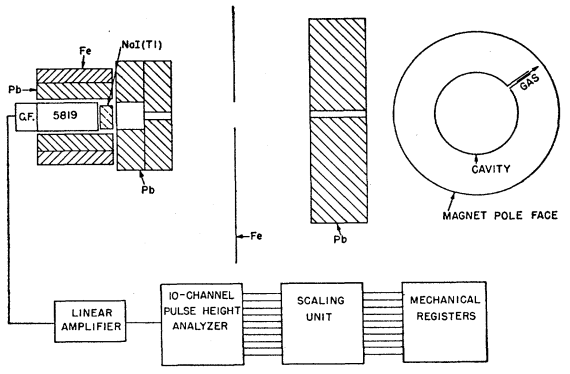

FIG. 1. Experimental arrangement.

Gamma rays due to positron annihilations are observed with a scintillation spectrometer in a direction at 90° to the direction of the magnetic field. Extensive lead shielding provided with a $\frac{1}{2}$ in. viewing hole is placed between the cavity and the crystal which is about 23 in. from the center of the magnetic field. Furthermore, an iron shield is connected so as to minimize stray flux from the magnet at the detector. The scintillation spectrometer uses a NaI(Tl) crystal and a 5819 photomultiplier tube, placed inside a lead and iron shield. The pulses from the photomultiplier are fed through a cathode follower and linear amplifier into a ten-channel differential pulse-height analyzer of conventional design using 1N38 Germanium diodes and thence into scalers and mechanical registers. Each channel of the analyzer can be expanded into 10 subchannels to obtain a detailed view of the energy spectrum. A subchannel spans an energy range of about 7.5 kev. Before each run the scintillation spectrometer is calibrated with a Na^{22} and a Cs^{137} source; during the course of an experiment ($\sim$12 hours) the calibration changes at most 2 subchannels.

4. GAMMA-RAY ENERGY SPECTRA

The observed energy spectra arise from several processes.

(1) Positron annihilation in the gas either as free positron-electron annihilation or as annihilation from the bound states of parapositronium or orthopositronium. The free positron-electron annihilation radiation will be a two-quantum process indistinguishable from the parapositronium decay.

(2) Positron annihilation from the walls of the container, which will be principally two-quantum annihilation.

(3) Degradation of the gamma rays arising from the above processes, due principally to Compton effect collisions in the gas or in the container walls.

The negative beta particles emitted by Cu^{64} will be completely absorbed in the container. There is a 1.34-

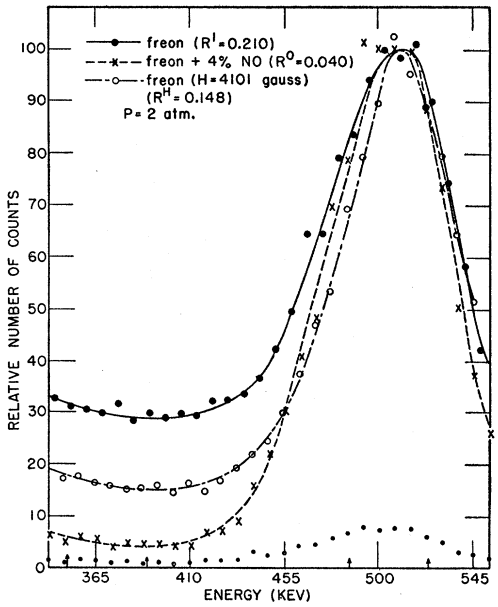

FIG. 2. Positron annihilation spectra: Number of counts *vs* γ-ray energy, normalized to 100 at the peak. Number of counts in vacuum with no magnetic field is indicated by small solid circles at bottom of graph. Peak region from 485 to 530 kev and valley region from 345 to 390 kev are indicated by arrows.

Mev gamma ray which occurs with the relative frequency (No. of γ)/(No. of β^+)=0.05 and will contribute to the background.

The magnetic quenching effect to be observed is a conversion of a certain amount of the three-quantum orthopositronium decay to two-quantum parapositronium decay, which will shift counts from the lower energy distribution characteristic of orthopositronium decay to the energy mc^2 characteristic of parapositronium decay. Compton degradation and the energy resolution of the apparatus tend to obscure this effect. Another complicating factor is a possible change in the background wall counts, and, in general, a change in the distribution of positron decays throughout the cavity when the magnetic field is changed. These factors will be discussed in detail later in the section on analysis of data.

The curves in Fig. 2 show some observed spectra. The solid curve drawn through the solid dots was observed with freon 12 (CCl_2F_2) in the cavity at a pressure of 2 atmospheres, and with no magnetic field present. The peak counting rate occurs for an energy of about 510 kev (mc^2). The counting rate in the valley region about 400 kev is contributed in part by degraded gamma rays from either two quantum or three quantum decays. The dashed curve drawn through the crosses was observed with the same conditions as for the previous curve with the exception that a small amount of NO gas was added. (The NO added contributed about 4 percent of the pressure of 2 atmospheres.) The large reduction in the valley to peak ratio, from about 0.210 to 0.040, caused by the introduction of the NO is interpreted as due to the conversion of all orthopositronium to parapositronium by collisions with NO molecules. Finally, the dot-dash curve through the open circles is observed with freon alone at a pressure of 2 atmospheres and with a magnetic field of 4101 gauss. The reduction in the valley to peak ratio compared to the ratio for curve 1 is interpreted as due to the conversion of $M=0$ orthopositronium annihilation from the three-quantum to the two-quantum decay, and is the static magnetic field quenching effect to be studied. All three curves were normalized to the same value at an energy of 510 kev.

5. ANALYSIS OF DATA

(5.1) Simplest Approach

To determine f, the fraction of orthopositronium atoms which decay by three-quantum annihilation, the following analysis is applied to spectra of the three types given in Fig. 2. The following formulas give the number of counts in the peak and valley regions in terms of the relevant parameters:

$$\begin{aligned}
P^1&=N_pg_p+N_ag_p+N_og_o,\\
V^1&=N_ph_p+N_ah_p+N_oh_o,\\
P^0&=N_pg_p+N_ag_p+N_og_p,\\
V^0&=N_ph_p+N_ah_p+N_oh_p,\\
P^H&=N_p{}^Hg_p+N_a{}^Hg_p+N_o{}^Hfg_o+N_o{}^H(1-f)g_p,\\
V^H&=N_p{}^Hh_p+N_a{}^Hh_p+N_o{}^Hfh_o+N_o{}^H(1-f)h_p,
\end{aligned} \tag{6}$$

in which the superscripts 1, 0, and H refer respectively to the conditions pure freon and no field, freon plus NO and no field, and freon and a field H. P is the number of counts in the peak of the energy spectrum which was arbitrarily taken to include counts in 6 subchannels which is an energy range of about 45 kev. V is the number of counts in the valley of the energy spectrum which was again taken to include counts in 6 subchannels. The quantities N_a, N_p, and N_o are, respectively, the number of free positron-electron annihilations, the number of parapositronium atom annihilations, and the number of orthopositronium atom annihilations in the region of the cavity viewed by the spectrometer. The quantity g_p is the average probability that an annihilation γ ray from a parapositronium decay will be counted in the peak region of the energy spectrum. This factor depends upon γ-ray degradation effects, the spatial distribution of annihilation events, the solid angle subtended by the detecting crystal, and the efficiency and resolution of the scintillation spectrometer. The average probability that an annihilation γ ray from a free positron-electron annihilation will be counted in the peak region is taken to be the same quantity, g_p. The quantity g_o is the average probability that a γ ray from an orthopositronium decay will be counted in the peak region. For the valley

TABLE I. γ-ray counts.

Condition		Peak counts	Valley counts	Ratio of valley counts to peak counts	$f(H)$
Case 1: Freon alone, no field					
	9:50 P.M.	13 594±0.9%	2798±1.9%	0.206	
	11:10 P.M.	12 856	2581	0.201	
	11:55 P.M.	12 316	2532	0.206	
	12:55 P.M.	11 476	2304	0.201	
	1:20 A.M.	11 269	2307	0.205	
	2:15 A.M.	10 815	2157	0.199	
	5:20 A.M.	9256	1953	0.211	
	6:20 A.M.	8595	1801	0.210	
	7:15 A.M.	8429	1818	0.216	
				Average=0.206	
Case H: Freon alone, field H					
H=1788 gauss	10:40 P.M.	19 784±0.7%	3296±1.7%	0.167	0.805
H=2572 gauss	11:30 P.M.	22 352	3546	0.159	0.734
H=3284 gauss	12:20 P.M.	24 927	3599	0.144	0.716
H=4101 gauss	1:40 A.M.	26 859	3912	0.146	0.675
H=5620 gauss	2:35 A.M.	30 524	4306	0.141	0.631
H=6590 gauss	3:50 A.M.	31 692	4261	0.134	0.588
H=6940 gauss	5:00 A.M.	29 681	4024	0.136	0.595
H=1140 gauss	6:00 A.M.	11 007	2091	0.190	0.933
Case 0: Freon plus NO, no field					
	8:05 A.M.	10 105±1%	474±4.6%	0.047	
	8:15 A.M.	10 052	437	0.043	
	8:25 A.M.		462	0.046	
				Average=0.0453	
No gas, no field[a]	1:55 A.M.	1084±3%	140±8.5%	0.129	
	7:40 A.M.	713	104	0.146	

[a] Note: Data on the condition no gas and magnetic field present were not taken on the run for which data are given in this table. However, data taken on other runs indicated that at a field of about 2500 gauss such a background reading has a number of counts in the valley about 3.5 percent of the gas plus field valley count and a number of counts in the peak about 0.7 percent of the gas plus field peak count.

region the corresponding probability factors are designated by h_p and h_o.

It is seen from the formulas that the same probability factors are used for the three different experimental conditions, which implies the assumption that the spatial distribution of the annihilations is the same in the three cases. Though it would be expected that this assumption is true for cases 1 and 0, it is not so apparently true for case H in which the motion of the positrons is greatly influenced by the presence of the magnetic field. Nevertheless, it is believed to be a good assumption, because the collimation of the γ-ray detection system is good enough so that only a small number of the counts arise from background wall counts in any of the cases, and spatial redistributions throughout the volume of the gas would not be expected to change the probability factors appreciably. It should be noticed that in case 0 the probability factors associated with orthopositronium atoms are taken to be g_p and h_p, which corresponds to the assumption that all orthopositronium atoms formed are quenched by collisions with NO molecules so that they decay by two-quantum annihilation. The assumptions have been implicitly made in the formulas (6) that the number of free positron-electron annihilations and the numbers of parapositronium and orthopositronium atoms formed are the same in the case of freon alone or of freon plus NO. However, it is not assumed that $N_p=N_p{}^H$, for example, since the presence of the magnetic field could change the number of positronium atoms formed in the active region of the gas. Finally, it is assumed for all three cases that $N_o=3N_p$ and $N_a=KN_p$, in which K is a constant; or that the relative number of orthopositronium to parapositronium atoms is the statistical weight factor in the three cases, and that the relative number of free positron-electron annihilations to parapositronium decays is the same in all three cases. Effectively, various of these assumptions imply that background counts arising from the annihilation of positrons in the cavity walls are neglected; this assumption will be discussed later in this section.

The experimental data give directly values for the P's and V's, and, with the assumptions mentioned above, Eqs. (6) can be solved for f to yield:

$$f=1/(1+y), \tag{7}$$

in which $y=\rho(R^1-R^H)/(R^H-R^0)$, where $\rho=P^1/P^0$, $R^1=V^1/P^1$, with analogous definitions for R^0 and R^H.[12]

Data from a typical run are shown in Table I. Each of the entries for number of counts corresponds to an observation time of 5 minutes and is the sum of the counts from 6 subchannels or from an energy range of about 45 kev. The percentage statistical accuracy is

[12] M. Deutsch, Massachusetts Institute of Technology Progress Report, August 31, 1951 (unpublished).

indicated for several of the readings. It is seen that for case 1 the statistical accuracy for the determination of the ratio of valley counts to peak counts is about 2 percent, and that the various determinations agree within expected statistical limits. For case H the counting rates are increased over those of case 1 by as much as a factor of 3 at high magnetic field values; this increase is due to the focusing of the positrons by the magnetic field in the center of the cavity which is the region viewed by the spectrometer. The statistical accuracy with which the ratios of valley counts to peak counts can be determined in case H is somewhat better than for case 1. The largest statistical uncertainty is encountered in case 0 because of the small number of valley counts; furthermore, these data were customarily taken at the end of a run when the source strength was weakest, because of the difficulty of cleaning out all the NO from the system in a reasonably short time once it had been admitted.

A few remarks must be made about the treatment of background wall counts which was employed to obtain the ratio of valley to peak counts given in the table. The method of analysis of the data assumed that all of the γ rays originate from annihilations in the gas and that none come from annihilations in the container walls. This assumption was essential because, otherwise, if wall annihilations occurred and if the number of wall annihilations relative to the number of gas annihilations varied with variation of the magnetic field, the shape of the energy spectrum would change with field for this reason alone. In actuality some counts are expected to arise from annihilations in the walls, so the problem is to subtract these counts from the peak and valley counts before beginning the analysis as presented. The data in Table I include peak and valley counts for the condition of no gas and no field. The majority of these counts arise from positron annihilations; only a small percentage of these counts would originate from the 1.34-Mev γ ray of Cu^{64} or external radiation sources (cosmic rays, laboratory contamination, etc.) as is clear from the energy spectrum for this case. The peak and valley counts in this case are about $\frac{1}{10}$ of the counts observed in case 1 when freon is present. The valley counts observed with no gas present are about $\frac{1}{3}$ of the valley counts observed in case 0 with freon plus NO, however, and the uncertainty as to the proper method of subtracting the wall background counts from the case 0 valley counts is an important source of error in this experiment. Peak and valley counts are also given in Table I for the condition of no gas but with a field present. At the higher fields these background counts amount only to about 3 percent of the case H counts.

The method of subtracting background wall counts consists in arbitrarily subtracting from the valley or peak counts observed in any case $\frac{1}{2}$ of the valley or peak counts observed with no gas and corresponding magnetic field. The motivation for this procedure is the remark that the background wall counts observed in the no gas case are certainly higher than would be expected when gas is present, because in the latter case the majority of the positrons will annihilate in the gas rather than in the walls. By subtracting $\frac{1}{2}$ of the no-gas counts an error of less than this subtracted number is expected. For cases 1 and H the corresponding error in valley-to-peak ratio is less than 1 percent; for case 0 it is less than 10 percent. The total uncertainty produced in f as a result of background counts is thus about 2 percent.

Values of $f(H)$ computed from these data by using Eq. (7) are shown in the last column. The quantity ρ is taken to be 0.82. The statistical counting accuracy to which the various values of $f(H)$ are determined is about $2\frac{1}{2}$ percent.

The magnetic field was measured either by the observation of the frequency of a nuclear resonance absorption for H or Li^7,[13] or by the use of a GE flip coil fluxmeter. Values of the field measured by nuclear resonances are quoted to four figures and those measured by the fluxmeter are quoted to three figures. In all cases the accuracy to which the field is measured at a particular region together with the homogeneity of the field (measured at times other than during a run) suffice to determine H over the region of interest to within at least 1 percent, which is quite adequate for this experiment.

A principal factor limiting the accuracy in the determination of $f(H)$ is the stability of the scintillation spectrometer. No temperature control of the NaI(Tl) crystal or of critical circuit components was provided. The circuit instabilities—due mainly to the characteristics of the 1N38 diodes which perform the pulse-height analysis function—were such that longer running times and hence higher counting rates would not have been too profitable. Furthermore the ambiguities in the treatment of background wall counts is of the order of the statistical accuracy.

Figure 3 includes a plot of all the experimentally determined values of f *vs* the magnetic field; in most

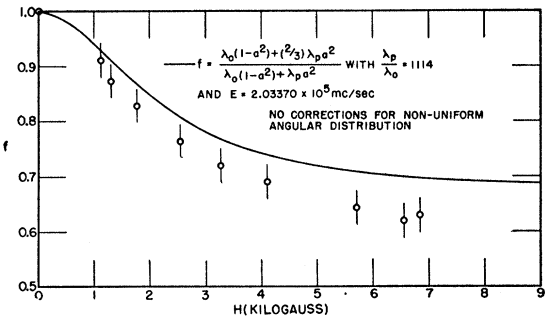

FIG. 3. Theoretical curve and experimental points for the fraction, f, of orthopositronium which decays by three-quantum annihilation as a function of the magnetic field. All experimental data were taken in freon at about two atmospheres pressure.

[13] R. V. Pound and W. D. Knight, Rev. Sci. Instr. **21**, 219 (1950).

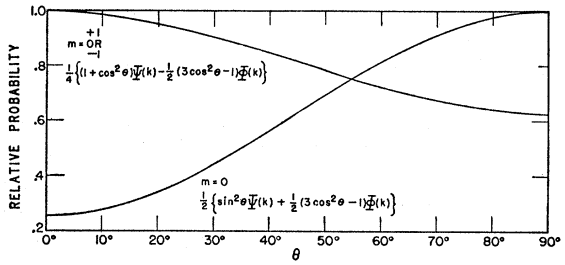

FIG. 4. Plot of Drisko's formulas for the relative probability of emission of annihilation quanta with energy kmc^2 by magnetic substates of orthopositronium. An average is taken over k between 0.71 and 0.79 as indicated by:

$$\int_{0.71}^{0.79}\Phi(k)dk\Big/\int_{0.71}^{0.79}\Psi(k)dk=0.229.$$

cases a point at a particular field is the average of several determinations of f made on independent runs. All data were taken in freon at about two atmospheres pressure. Errors which include counting statistics and background uncertainties are indicated for the points. The solid curve is a plot of the theoretical quenching formula (3′); the theoretical value of $\lambda_p/\lambda_o(=1114)$ is taken and the experimental value of the energy separation between parapositronium and orthopositronium is used. It is quite clear that the experimental points fall consistently below the theoretical curve. Additional data of somewhat less accuracy than the above were obtained with SF_6 and argon at two atmosphere pressure, and exhibited the same behavior as shown for the data of Fig. 3.

(5.2) Effect of Angular Distribution of Annihilation Radiation

The explanation for this discrepancy lies in the difference in the angular distribution of the annihilation quanta from the different magnetic substates of orthopositronium. Figure 4 is a plot of Drisko's formula for the relative probability of emission of annihilation quanta from the magnetic substates of orthopositronium as a function of angle for the energy range defined by $0.71\leq k\leq 0.79$ which corresponds to the valley region of our energy spectra. It is seen that there is a higher probability for the emission of an annihilation quantum in a direction at 90° to the field direction from the $M=0$ state of orthopositronium than from the $M=\pm 1$ substates. Such an angular distribution is of the right kind to explain the observed discrepancy, because by quenching the three-quantum decay of $M=0$ orthopositronium by a magnetic field (the decay of the $M=\pm 1$ substates are unaffected by the presence of the field), one removes more than $\frac{1}{3}$ of the triplet annihilation quanta observed by the scintillation crystal which is viewing at 90° to the direction of H.

Hence the formulas (6) used for the analysis of the experimental data must be modified to recognize this angular distribution effect, as indicated in the following equations:

$$\begin{aligned}
P^1&=N_pg_p+N_ag_p+\tfrac{1}{3}N_oG_0g_o+\tfrac{2}{3}N_oG_1g_o,\\
P^0&=N_pg_p+N_ag_p+N_og_p,\\
P^H&=N_p{}^Hg_p+N_a{}^Hg_p+\tfrac{1}{3}N_o{}^HFG_0g_o\\
&\qquad+\tfrac{1}{3}N_o{}^H(1-F)g_p+\tfrac{2}{3}N_o{}^HG_1g_o.
\end{aligned}\tag{8}$$

There will be corresponding expressions for V^1, V^0, and V^H in which there will appear H_0 and H_1 instead of G_0 and G_1. In these formulas, for example, the product G_0g_o replaces g_o of Eqs. (6), and the product has the same meaning of average probability that an annihilation gamma ray from $M=0$ orthopositronium three-quantum decay shall be counted in the peak region. There is a corresponding interpretation for the product G_1g_o. The G's are introduced to take into account the difference in these probabilities for the different magnetic substates. It is permissible to set $G_0=1$, and then G_1 is the probability that a decay γ ray from a three-quantum decay of $M=+1$ or $M=-1$ orthopositronium will be emitted in a direction at 90° to the magnetic field (the observation direction) relative to the same probability for $M=0$ orthopositronium. This quantity can be evaluated from Eq. (4) as:

$$G_1=\frac{\Psi(k)+\frac{1}{2}\Phi(k)}{2[\Psi(k)-\frac{1}{2}\Phi(k)]}.\tag{9}$$

Its average value over the peak energy range from 470 to 510 kev is evaluated as $G_1(=0.62)$. In an analogous fashion take $H_0=1$ and evaluate $H_1(=0.80)$ over the valley energy range $k=0.71$ to 0.79 or from 360 to 400 kev. (The difference between these energy intervals and those shown in Fig. 2 is not significant in the evaluation of the G's and H's.)

$$\begin{aligned}
f&=1/(1+Z),\\
Z&=\rho(R^1-R^H)/(R^H-R^0+C),\\
C&=\frac{2(1-H_1)}{1+2H_1}(R^1\rho-R^0K')+\frac{2(1-G_1)}{1+2G_1}R^H(K'-\rho),\\
K'&=(1+K)/(4+K).
\end{aligned}\tag{10}$$

It will be noticed that if there were no difference in the angular distribution of the annihilation radiation from the different magnetic substates of orthopositronium, then $G_1=H_1=1$ and the formulas for f would be identical to Eq. (7).

In the above expression for f there appears the quantity K which is the relative number of free positron-electron annihilations to parapositronium decays. The quantity K can be determined in a manner similar to that used by Pond.[14] The increase in the number of peak counts between case 1 and case 0 (i.e., caused by the addition of a small amount of NO) is interpreted as due to the conversion of all orthopositronium atoms

[14] T. A. Pond, Phys. Rev. 85, 489 (1952).

to parapositronium atoms. Assuming equal probabilities for the formation of parapositronium and any magnetic substate of orthopositronium, one third of this observed increase will be the number of decays due to parapositronium atoms in case 1. The number of free positron-electron annihilations is then the number of peak counts in case 1 minus the number of parapositronium decays in case 1 as just determined and minus the fraction of orthopositronium decays which would appear among the peak counts in case 1 (this fraction is estimated from the γ-ray energy distribution curve of Ore and Powell[3]). An estimate of background wall counts can be made by observing the peak counts in the absence of gas in the cavity. As seen from Table I, the background counts amount to only about 10 percent of the gas counts; background counts can be subtracted from the peak counts of case 1 before the indicated calculation is performed. By such a calculation it is found that $K=16$ for freon; this value is the same magnitude as the values found by Pond for other gases.

By using this value for K, values of $f(H)$ can be computed for data like that given in Table I. It should be emphasized that, although K is not accurately known, it is sufficiently well known for the purpose at hand. The quantity K appears only in the part of formula (10) which represents the correction due to the angular distribution effect. Furthermore, K appears in the combination $K'=(1+K)/(4+K)$ which changes only slowly with change in K, when K is substantially greater than 1, as is the case here. Solutions were obtained for other values of K within a reasonable range and also for other reasonable values of G_1 and H_1; within statistical experimental errors these various solutions did not differ significantly.

Figure 5 shows the data of Fig. 4 recomputed to take into account the angular distribution effect. Statistical errors are indicated on the points. The experimental points are regarded as in adequate agreement with the theoretical quenching curve.

6. DETERMINATION OF RATIO OF DECAY RATES OF PARAPOSITRONIUM AND ORTHOPOSITRONIUM

For the determination of the ratio λ_p/λ_o from the experimental data, the following procedure is used. The correct form for the theoretical quenching curve is taken to be formula (3′). The energy separation between orthopositronium and parapositronium is taken as 2.0337×10^5 Mc/sec from the radio-frequency experiment, and the magnetic field was measured so that a is determined. Hence a value of f, calculated from the experimental data as described for Fig. 5, will yield a corresponding value of λ_p/λ_o, which will be designated Λ. Λ is given by $\Lambda=(1-F)/AF$, in which $A=a^2/(1-a^2)$. It will be appreciated that the greatest accuracy in the determination of Λ will come from experimental points which fall on the rapidly changing part of the magnetic quenching curve, and hence greatest weight must be given to these points. Formally, the uncertainty in the ratio Λ can be related to the uncertainty in the determination of $F(H)$ by the expression:

$$\Delta\Lambda=\Delta F/AF(F+\Delta F) \text{ in which } f=(2+F)/3.$$

For values of H above 5500 gauss, F is less than about 0.12, and, indeed, is of the order of ΔF so that $\Delta\Lambda$ can be large; and the weighting factor, which would be proportional to $1/(\Delta\Lambda)^2$, is considerably less than $\frac{1}{10}$ for all such points, relative to points taken at lower fields. Actually only experimental data for fields below about 4000 gauss are used for the determination of Λ. The value of Λ depends very critically upon the value of f so that a 2 percent error in f leads to an error in Λ of about 20 percent.

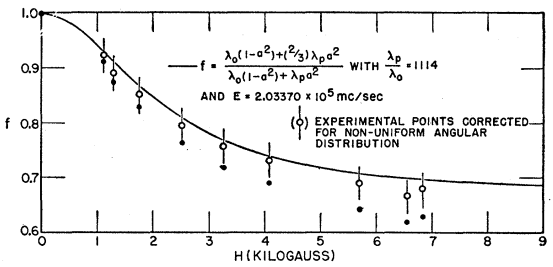

FIG. 5. Similar to Fig. 3 with the inclusion of corrections for the angular dependence of the annihilation radiation from the magnetic substates of orthopositronium. Solid dots are uncorrected points of Fig. 3.

A determination of Λ, to provide a least squares fit to the theoretical formula (3′) for f, yields $\Lambda=1302$ with a standard deviation of ±100. An error of ±15 percent is assigned as a probable error to account principally for statistical counting errors and uncertainties in the treatment of the background. This value is in reasonable agreement with the theoretical value of 1114.

If the effect of collisions on the quenching of the decay of orthopositronium were appreciable under the experimental conditions, a correction would need to be made in the above procedure for computing λ_p/λ_o. Deutsch[15] has studied the rate of collision quenching of orthopositronium in freon and quotes a value of 0.3×10^6 sec^{-1} atmos^{-1}. Deutsch remarks, however, that this value is "rather uncertain," and it appears from the published data that the collision quenching may well be zero within the experimental error. If the value for collision quenching of 0.6×10^6 sec^{-1} at 2 atmos is used, our data would yield $\lambda_p/\lambda_o=1410$ under the assumption that collision quenching is the same for all magnetic substates of orthopositronium. In view of the uncertainty of the collision quenching rate, no correction is made for this effect in the value of $\lambda_p/\lambda_o(=1302)$ quoted above.

A more accurate measurement of λ_p/λ_o would require a considerable reduction in vacuum background counts, a greater statistical counting accuracy, improved sta-

[15] M. Deutsch, Phys. Rev. 83, 866 (1951).

bility of the scintillation spectrometer, a better knowledge of the collision quenching rate, and perhaps a more careful calculation of the angular distribution correction as regards averaging over the peak and valley energy regions. Furthermore, it was observed upon numerous occasions that the valley-to-peak ratio in freon would decrease with time over a period of several hours, suggesting some mechanism which was quenching positronium—perhaps a buildup of a gas impurity or of ionization due to the radioactivity. By sufficiently frequent changes of the gas this effect was believed to introduce no error in the determination of f to the order of 2 percent, but for any greater accuracy the effect would be troublesome.

It might be worth mentioning that one motivation for a more precise measurement is that the presence of $2S$ orthopositronium might be exhibited as an apparent high value of the ratio λ_p/λ_0. Indeed a 10^{90} increase in the apparent value of λ_p/λ_0 as calculated in this paper would result if the relative number of $2S$ positronium atoms to $1S$ positronium atoms is of the order of 1 to 30. A higher precision measurement of λ_p/λ_o is being planned, and a measurement of the magnetic quenching effect at $\theta=0°$ is under way.

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Rotational Excitation by Slow Electrons. II*

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Theoretical cross sections for the rotational excitation of homonuclear molecules by slow electrons are applied to calculation of the fractional energy loss per collision (λ) in H_2. The theoretical losses are not more than $2.5(2m/M)$ and except at the lowest energies studied ($\sim$0.1 ev) are smaller than observed.

It would be desirable to have more direct experimental evidence of rotational excitation. For this reason we have calculated λ at 77°K in pure para-hydrogen and in normal hydrogen at that temperature. At electron energies $\sim$0.075 ev, the two λ's should differ by about 50 percent. Similarly, because of the altered rotational distribution, λ for deuterium differs from λ for H_2. Such differences, if observed, could hardly be accounted for on any other basis than rotational excitation.

I. INTRODUCTION

In a previous paper[1] cross sections were calculated for the rotational excitation of homonuclear diatomic molecules by very slow electrons. These cross sections were used to compute λ, the average fractional energy loss per collision. In nitrogen it was found that, at energies below the vibrational threshold, excitation of rotational levels easily accounted for the observed excess of λ' (λ in units of $2m/M$) above the purely elastic value of unity. The calculations of I are here applied to hydrogen for which recent swarm measurements are also available.[2]

In Fig. 1 are compared the theoretical and observed values of λ' for H_2. The notation and procedure used to compute the curves are the same as in I. In hydrogen,[3,4] $B=7.5\times10^{-3}$ ev, $Q=0.393$ in units of ea_0^2, and we use a total cross section[5] $\sigma_t=13.5\pi a_0^2$, independent of energy. The calculations apply to H_2 at 290°K, with a Boltzmann distribution of rotational levels. The theoretical values of λ' are seen to be smaller than the experimental values even at average energies well below the vibrational threshold (0.54 ev). Our estimate of the energy loss due to rotational excitation accounts for only about half the observed excess of λ above $2m/M$. It is unlikely that these departures between the theory and observations are wholly due to vibrational excitation by higher energy electrons in the swarm.[6] The significance of the discrepancy between the curves of Fig. 1 is difficult to

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[2] R. W. Crompton and D. J. Sutton, Proc. Roy. Soc. (London) **A215**, 467 (1952).

[3] G. Herzberg, *Spectra of Diatomic Molecules* (D. Van Nostrand and Company, New York, 1950), second edition, p. 532.

[4] N. J. Harrick and N. F. Ramsey, Phys. Rev. **88**, 228 (1952).

[5] L. J. Varnerin, Jr., Phys. Rev. **84**, 563 (1951).

[6] For average electron energies of about 0.1 ev and assuming a Maxwellian distribution in the swarm, the magnitude of the vibrational excitation cross section required to account for the difference between the theoretical and experimental curves of Fig. 1 is $\sim0.3\pi a_0^2$. This probably is an underestimate of the required cross section since the tail of the electron energy distribution is expected to fall off more rapidly than Maxwellian. T. Holstein, Phys. Rev. **70**, 367 (1946); D. Barbiere, Phys. Rev. **84**, 653 (1951). Little is known about the value of the vibrational cross section near threshold, but the measured values at higher energies are much smaller than $0.3\pi a_0^2$. H. Ramien, Z. Physik **70**, 353 (1931); Chao, Wang, and Shen, Science Record (China) **2**, 358 (1949). Moreover, none of the somewhat disparate theoretical estimates are nearly this large. H. S. W. Massey, Trans. Faraday Soc. **31**, 556 (1935); T. Y. Wu, Phys. Rev. **71**, 111 (1947); P. M. Morse, Phys. Rev. **90**, 51 (1953); T. R. Carson, Proc. Phys. Soc. (London) **A67**, 909 (1954).