Magnetic Quenching of Positronium in Solids and Positron Helicity*

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(Received June 11, 1962)

The magnetic quenching of the positronium decay was investigated in Teflon and Lucite as a function of the magnetic field up to 18 600 G by differentiating the triplet from the singlet state on the basis of annihilation time spectra. The experimental results strictly agree with the theoretical provisions. Using the theoretical value of the ground-state splitting our results led to the mean life of the positron in singlet positronium with its own electron equal to $(1.23\pm0.08)\times10^{-10}$ sec and to $(1.21\pm0.11)\times10^{-10}$ sec, respectively, for Teflon and Lucite. The polarization of the positrons just prior to capture was deduced by measuring the asymmetry in the counting rate due to the different positron's relative spin population of the ground-state positronium in a magnetic field. We found $P=+(0.75\pm0.11)(\bar{v}/c)$ and $+(0.73\pm0.13)(\bar{v}/c)$, respectively, in Teflon and Lucite, where $(\bar{v}/c)$ is the mean value of the positron longitudinal polarization over the β spectrum.

I. INTRODUCTION

As is well known, the states of positronium show no linear Zeeman effect. However, there is a second-order magnetic effect which gives rise in the ground state to a displacement of the two substates with $m=0$ with respect to the unperturbed position; furthermore, the two states become mixed.[1] This leads to possible two-quantum decay of the $m=0$ ortho state and thereby to a diminution of its lifetime which is progressively replaced, with increasing magnetic field, by the much shorter lifetime of the para state.

The effect has been investigated in gases by several authors[2–4] who, by measuring the quenching of the orthopositronium three-photon annihilation in a static magnetic field, achieved a good agreement with the theory. A further refinement of these experiments has been obtained by introducing radio-frequency transitions between the ortho states[5,6]; in this way the hyperfine structure splitting was determined with so high an accuracy as to provide a confirmation of the theory to order of α^3 Ry.

As far as the annihilation in solids is concerned, some of the available measurements give only qualitative informations on the magnetic effect; in a few cases they demonstrate consistency with the expectation but do not permit quantitative agreement. Experiments in solids on the quenching of the three quantum annihilation of orthopositronium by a magnetic field have been performed by Telegdi *et al.*[7] The results of this measurement are of remarkable interest as far as concerns the behavior of positron annihilation in amorphous insulators, but they do not give the functional dependence of the effect from the magnetic field. Measurements of the angular correlation of the two annihilation quanta have been carried out by Warshaw[8] and by Page and Heinberg.[9] These authors found that in some materials where positronium is thought to be formed, the narrow component which by itself is a strong evidence of parapositronium, is enhanced in the presence of a magnetic field. Delayed coincidence measurements were performed in polystyrene by Freytag and Ziock[10] who put in evidence the influence of a magnetic field on the orthopositronium lifetime.

It should be emphasized that the interpretation of magnetic quenching in solids differs from that in gases for the following essential reason: The orthopositronium decays by "pick-off"[11,12] with a lifetime much shorter than the "natural" lifetime for three-photon decay. As a consequence three-photon magnetic quenching measurements enable us to determine the parameter $\lambda_s/\gamma(\hbar\omega_0)^2$, λ_s being the annihilation rate of positronium in the singlet state, γ the rate of "pick-off" annihilation, and $\hbar\omega_0$ the ground-state splitting, while in gases the derived parameter is $\lambda_s/\lambda_3(\hbar\omega_0)^2$, λ_3 being the well-known "natural" three-photon decay rate. Unfortunately, the "pick-off" rate γ cannot at present be calculated, so that the quoted quenching experiments are unable to test theoretical predictions unless the results of independent lifetime measurements are utilized. But this last possibility is more apparent than real, in view of the fact that the dependence of "the pick-off" lifetime on several factors, like temperature, pressure, phase, and processing of the material, makes the results of different authors hardly comparable among each other.

The basic feature of the experiment which will be described in this paper was to differentiate the triplet from the singlet state through annihilation time spectra. This procedure allows a quantitative investigation of magnetic quenching in solids without implying any

* Research supported in part by the Consiglio Nazionale delle Ricerche.

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accurate knowledge of the "pick-off" lifetime. Furthermore, this is a more powerful method because the counting times which are required for gaining reliable information, are very short if compared with those typical of angular correlation and three-photon decay experiments.

The method of time-spectra measurement used by us gives, in addition, the opportunity to obtain the helicity of positrons which had slowed to near zero energy: A further differentiation between singlet and triplet states can be made on account of the fact that the formation of either the singlet or triplet state in a magnetic field is favored, depending on whether the incoming positrons are parallel or antiparallel to the field. Page *et al.*[13,14] made use of this effect for the first time, by measuring the preferential narrowing of two-photon angular correlation due to the preferential capture into triplet state of positrons with spin opposite to the direction of magnetic field. The method used in the present paper is particularly suitable for obtaining information on the depolarization of positrons in matter if one assumes that the helicity of positrons just after emission is equal to $+v/c$, as required by the two-component neutrino theory. Direct measurements of positron helicity, while compatible with this value, are neither extensive nor accurate to more than 15%.[15]

II. THEORY OF THE EXPERIMENT

The results of the present section are mostly known but are reported in what follows for the sake of convenience.

At zero field a pure singlet state of free-space positronium annihilates with emission of two quanta traveling in opposite directions, each having an energy of mc^2 with a decay rate $\lambda_s=8.0\times10^9$ sec^{-1}. A pure triplet state in free space annihilates with emission of three coplanar quanta having a continuous energy distribution up to a maximum of mc^2, with a decay rate $\lambda_3=7.2\times10^6$ sec^{-1}. In solids an additional possibility of decay arises from the annihilation with an electron belonging to one of the surrounding atoms ("pick-off" annihilation); as the "pick-off" decay rate (a few times 10^8 sec^{-1}) is much lower than λ_s and greater than λ_3, the ortho state decays with a rate $\lambda_t=\lambda_3+\gamma$ typical of the matter, while the para state lifetime is hardly changed by the presence of the matter.

In presence of a magnetic field the $m=\pm1$ states of orthopositronium are still pure triplet states and their lifetime is left unchanged. On the contrary, the $m=0$ ortho state is perturbed by the field in such a way as to contain an admixture of singlet state; the same happens to the $m=0$ para state which becomes mixed. On the basis of the perturbation theory it is easy to deduce the eigenfunctions for the mixed states:

$$\Phi_2=\frac{1}{[2(1+y^2)]^{1/2}}[u_2-yu_4]\times\exp\left[-i\left(\frac{W}{\hbar}+\omega_0+\frac{M_z^2}{\hbar^2\omega_0}\right)t-\tfrac{1}{2}\lambda_t't\right], \quad (1)$$

$$\Phi_4=\frac{1}{[2(1+y^2)]^{1/2}}[yu_2+u_4]\times\exp\left[-i\left(\frac{W}{\hbar}-\frac{M_z^2}{\hbar^2\omega_0}\right)t-\tfrac{1}{2}\lambda_s't\right], \quad (2)$$

where u_2 and u_4 are the spatial and spin-dependent parts of the unperturbed eigenfunctions of the ortho state with $m=0$ and of the para state, respectively; $W=e^2/2a$ is the eigenvalue of the ground state; $\hbar\omega_0$ is the ground-state hyperfine structure splitting; and $M_z=(e\hbar/2mc)H_z$ is the only nonvanishing matrix element of the perturbation Hamiltonian.

The physical interpretation is simple: The perturbed ortho state contains an amount of pure singlet equal to $y^2/(1+y^2)$, where $y=[(1+x^2)^{\frac{1}{2}}-1]/x$ and $x=4M_z/\hbar\omega_0$ and decays with a rate

$$\lambda_t'=(y^2\lambda_s+\lambda_t)/(1+y^2);$$

correspondingly, the perturbed para state contains an amount of pure triplet state equal to $y^2/(1+y^2)$ and decays with a rate

$$\lambda_s'=(y^2\lambda_t+\lambda_s)/(1+y^2).$$

Consequently, the contributions to the two-quantum annihilations during the time dt given by the unperturbed states of orthopositronium and by the perturbed one, are respectively proportional to

$$\tfrac{2}{3}\lambda_t e^{-\lambda_t t}(\gamma/\lambda_t)dt,$$

where γ is the "pick-off" decay rate, and to

$$\tfrac{1}{3}\lambda_t' e^{-\lambda_t' t}(\gamma'/\lambda_t')dt,$$

where

$$\gamma'=(y^2\lambda_s+\gamma)/(1+y^2).$$

Similar expressions are obtained for the contributions given only by three quantum events. If one observes two-quantum decays and a fraction f of three-quantum decays arising from those atoms which have survived the time t_1, the number n of detected annihilations is

$$n\propto\tfrac{2}{3}e^{-\lambda_t t_1}\frac{\gamma+f\lambda_3}{\lambda_t}+\tfrac{1}{3}e^{-\lambda_t' t_1}\frac{\gamma+y^2\lambda_s+f\lambda_3}{\lambda_t+y^2\lambda_s}. \quad (3)$$

We are now able to define the quenching R (the quantity which will be measured) as the ratio between the detected annihilations with and without field. R

[13] L. A. Page and M. Heinberg, Phys. Rev. **106**, 1220 (1957).
[14] L. A. Page and F. E. Obenshain, Bull. Am. Phys. Soc. **2**, 260 (1957).
[15] L. Grodzins, *Progress in Nuclear Physics* (Pergamon Press, New York, 1959), Vol. 7, p. 165.

turns out to be

$$R=\frac{1}{3}\left\{2+\alpha\exp\left[-\frac{y^2}{1+y^2}(\lambda_s-\lambda_3)t_1\right]\right\}, \quad (4)$$

where

$$\alpha=\left[1-\frac{\lambda_3}{\lambda_t+y^2\lambda_s}(1-f)\right]\frac{1}{1-(\lambda_3/\lambda_t)(1-f)}.$$

In the preceding considerations we did not take into account any effect connected with the helicity of positrons, owing to the fact that we intended firstly to measure the quenching R by using random directions of positron emission with respect to magnetic field. However, if we utilize a positron beam exhibiting nonzero spin polarization with respect to an "average" direction of emission, the matter must be reconsidered.

As far as the decay rates are concerned, it should be emphasized that they depend on the square of the field strength and are, consequently, independent on field direction. On the contrary the positron's relative spin populations are affected by the field which favors the formation of singlet or triplet state for incoming positrons parallel or antiparallel to the field. If we rewrite the eigenfunctions of the perturbed states (1) and (2), emphasizing the spin-dependent parts,[15] we have, for the magnetic field pointing up:

$$\Phi_2\propto(\uparrow\downarrow)(1-y)+(\downarrow\uparrow)(1+y),$$
$$\Phi_4\propto(\uparrow\downarrow)(1+y)+(\downarrow\uparrow)(1-y),$$

where the first arrow indicates the positron spin.

It follows that the spin populations for the positronium states in a magnetic field up (i.e., parallel to the incoming positrons) are:

$$\tfrac{1}{4}(1\pm P)$$

for $J=1$, $m=\pm1$,

$$\tfrac{1}{8}[(1-\epsilon)(1+P)+(1+\epsilon)(1-P)]$$

for the perturbed "ortho state," and

$$\tfrac{1}{8}[(1+\epsilon)(1+P)+(1-\epsilon)(1-P)]$$

for the perturbed "para state." P indicates the polarization degree of the positron beam and its sign must be reversed for field down (i.e., antiparallel to the incoming positrons); ϵ is the quantity $2y/(1+y^2)$.

Then Eq. (3) is replaced by the two equations:

$$n_{\rm up}\propto\tfrac{1}{3}e^{-\lambda_t(t_1-t_2)}\frac{\gamma+f\lambda_3}{\lambda_t}+\tfrac{1}{6}(1-\epsilon P)e^{-\lambda_{t'}(t_1-t_2)}\frac{\gamma+y^2\lambda_s+f\lambda_3}{\lambda_t+y^2\lambda_s}, \quad \text{(field up)}$$

$$n_{\rm down}\propto\tfrac{1}{3}e^{-\lambda_t(t_1-t_2)}\frac{\gamma+f\lambda_3}{\lambda_t}+\tfrac{1}{6}(1+\epsilon P)e^{-\lambda_{t'}(t_1-t_2)}\frac{\gamma+y^2\lambda_s+f\lambda_3}{\lambda_t+y^2\lambda_s}, \quad \text{(field down)}$$

where we have taken into account only the atoms which survive longer than t_1 but less than t_2.

Once more we can define a quantity suitable for measurement, viz., the relative difference $2\Delta/\Sigma$ between the decay numbers for field up and field down:

$$2\frac{\Delta}{\Sigma}=2\frac{n_{\rm up}-n_{\rm down}}{n_{\rm up}+n_{\rm down}}=-2\epsilon P\frac{\alpha e^{-(\lambda_{t'}-\lambda_t)(t_1-t_2)}}{2+\alpha e^{-(\lambda_{t'}-\lambda_t)(t_1-t_2)}}. \quad (5)$$

III. EXPERIMENTAL METHOD

As our positron source we used Na^{22} which is customary for investigations on annihilation. Samples were prepared by evaporating one drop of high specific activity $Na^{22}Cl$ solution directly on thicknesses of Teflon and Lucite; these were covered with identical thickness of the same material when investigating the magnetic quenching, and with aluminum foil when investigating the positron helicity. In this last case the aperture angle of the beam of positrons forming positronium is limited to only 2π.

The γ rays were detected by means of two counters consisting of plastic scintillators NE102, $1\frac{1}{2}$-in. diam, 1 in. thick, or $1\frac{1}{2}$-in. diam, 2 in. thick, viewed by RCA 6342A photomultipliers through a 10-cm polyvinyltoluene light pipe. Two mu-metal cylindrical magnetic shields enveloped the phototubes. One counter was biased to accept only the pulses belonging to the Compton edge of the 1.28-MeV γ rays and the other was biased to accept only the Compton edge due to the annihilation radiation. The time distribution spectrum of the annihilation radiation with respect to the 1.28-MeV γ rays was analyzed by means of a time-to-height converter[16,17] and a 200-channel pulse analyzer. The prompt resolution curve (very approximately a Gaussian) has a full width at half-height of 8×10^{-10} sec and 10×10^{-10} sec, respectively, for the two different pairs of scintillators.

The magnetic field was produced by a magnet suitably designed for our purposes. It consists of magnetizing coils wound on a cylindrical iron core and completely surrounded by iron; the gap is located in the middle section. The uniformity of the field in the region useful for annihilation is certainly better than 0.7%. The two scintillators are placed at 180° near to the gap in order to obtain a detection efficiency as high as possible and the light pipes come out of the magnet through two windows. The stray field turns out to be so weak that it does not affect the photomultiplier at all. We have ascertained, through controls made during the whole set of measurements, that with field on, off,

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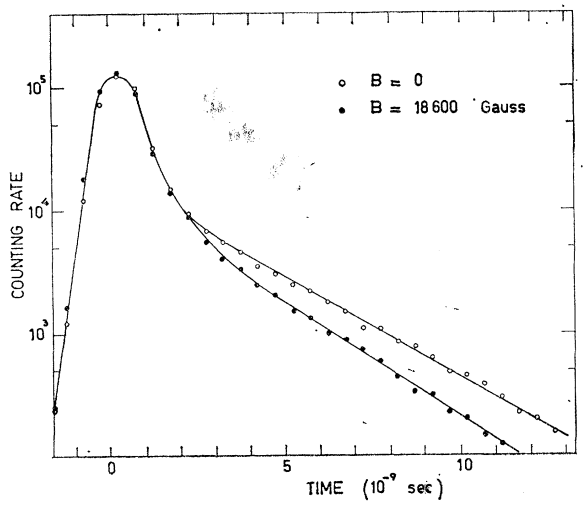

FIG. 1. Time spectra of annihilation radiation in Teflon with and without magnetic field. For convenience only one-half of the experimental points are marked.

or reversed, the centroid of the prompt resolution curve shifts less than 2×10^{-12} sec.

The time spectra of the annihilation radiation in the magnetic quenching experiments were recorded by storing it in the first hundred channels, while in the second hundred channels were stored the prompt pulses from Co^{60}. The set of measurements with zero field were distributed along the measurements with different values of magnetic field. A suitable distribution of recording times was selected in order to minimize any instrumental drifts.[18] In the helicity measurements the two hundred channels stored alternatively the time spectra with field up and down.

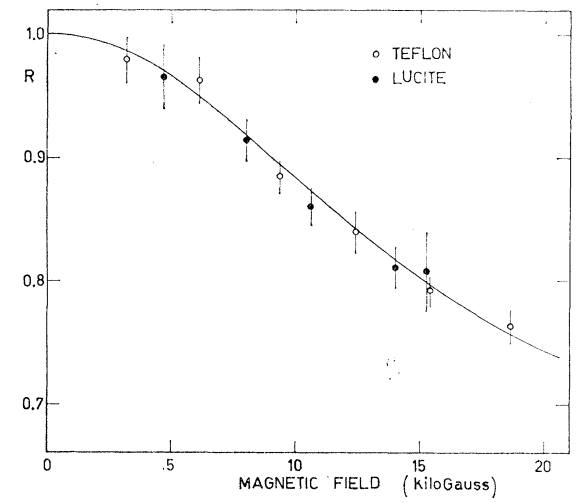

FIG. 2. Relative intensity of the long-lived positronium component with and without magnetic field.

[18] A. Bisi, A. Fasana, E. Gatti, and L. Zappa, Nuovo cimento 22, 266 (1961).

TABLE I. Ratio between the detected annihilations with and without field: measured and predicted values.

B (G)	$R_{measured}$ Teflon	Lucite	$R_{predicted}$
3200	0.979±0.017		0.985
4670		0.966±0.026	0.969
6100	0.963±0.019		0.950
8000		0.914±0.015	0.920
9350	0.885±0.013		0.897
10 600		0.860±0.012	0.875
12 400	0.839±0.015		0.844
14 000		0.811±0.015	0.819
15 300		0.808±0.033	0.800
15 400	0.793±0.010		0.799
18 600	0.764±0.012		0.758

The instant of time t_1 was selected as that for which the short time component contribution is not appreciable. The instant t_2 was chosen so as to have a counting rate statistically still significant. For the quenching experiment, t_1 was equal to 2.9×10^{-9} sec both for Teflon and Lucite; the contribution to the annihilation number from those atoms which have survived t_2, a small fraction of the total, was estimated from the spectra. For the helicity measurement, t_1 was taken equal to 2.9×10^{-9} sec for Teflon and 2.5×10^{-9} sec for Lucite; t_2 was 8.4×10^{-9} sec in the first case and 6.0×10^{-9} sec in the second one.

IV. RESULTS AND DISCUSSION

A. Magnetic Quenching

A typical spectrum of annihilation radiation in Teflon, which shows at a glance the effect of the magnetic quenching is reported in Fig. 1 for one value of the field. Numerical data are collected in Table I and fit strictly the theoretical curve as shown in Fig. 2. The curve is calculated on the basis of Eq. (4) taking into account the following considerations.

As mentioned in Sec. I, $\lambda_t=\lambda_3+\gamma$, being the three-quantum and the "pick-off" decay competitive processes for orthopositronium; similarly, $\lambda_s=\lambda_{s0}+\gamma$ being annihilation with its own electron and "pick-off" decay competitive processes for parapositronium. With the reasonable assumption that the "pick-off" annihilation rate is the same for the triplet and singlet states, Eq. (4) becomes

$$R=\frac{1}{3}\left\{2+\alpha\exp\left[-\frac{y^2}{1+y^2}(\lambda_{s0}-\lambda_3)t_1\right]\right\}. \qquad (6)$$

λ_{s0} indicates the decay rate of the positron in parapositronium with its own electron; one should expect its value to be slightly different from that typical of free-space parapositronium. As we do not know the

TABLE II. Relative difference $2\Delta/\Sigma$ (measured and predicted) between the annihilation numbers for field up and down.

	B (G)	t_1 (10^{-9} sec)	t_2 (10^{-9} sec)	$(2\Delta/\Sigma)$ predicted with $\eta=1$	$(2\Delta/\Sigma)$ measured	η
Teflon	12 500	2.90	5.50	-4.89×10^{-2}	$-(3.65\pm0.56)\times10^{-2}$	0.75
Lucite	13 000	2.50	3.50	-4.82×10^{-2}	$-(3.51\pm0.61)\times10^{-2}$	0.73

wave functions of positronium in solids, Eq. (6) was calculated by adopting for λ_{s0} and $\hbar\omega_0$ their free-space values. The λ_t values (3.88×10^8 sec^{-1} and 6.45×10^8 sec^{-1} for our samples of Teflon and Lucite, respectively) were taken off from the results of our previous measurements.[18] The fraction f of three-quantum decay was estimated to be equal to 0.54 on the basis of the known continuous three-quantum spectral shape and of its detection efficiency in our apparatus. The factor α turned out to be different from unity less than a few times 10^{-3} and so little sensitive to the material in which annihilation takes place that one cannot appreciate from the graph of Fig. 2 the difference between the curves for Teflon and Lucite.

From a comparison of the measured and predicted values of the quenching it is possible to extract some quantitative informations. Keeping for the hyperfine splitting $\hbar\omega_0$ the theoretical value 8.3462×10^{-4} eV, we find with a least-squares adjustment λ_{s0} (Teflon) $=(8.13\pm0.53)\times10^9$ sec^{-1} and λ_{s0}(Lucite) $=(8.27\pm0.75)\times10^9$ sec^{-1}. If this procedure is reversed we obtain with λ_{s0}(free-space) $=8.0\times10^9$ sec^{-1} the hyperfine splitting $\hbar\omega_0=(8.24\pm0.32)10^{-4}$ eV and $(8.17\pm0.45)10^{-4}$ eV for Teflon and Lucite, respectively.

It is straightforward to conclude that in solids (1) the long-lived component in annihilation spectra is to be attributed to the positronium atom in the triplet state, (2) the positronium atom is characterized by a hyperfine structure splitting and a decay rate in the singlet state, which differ from those in free space by certainly less than a few percent.

B. Positron Helicity

All the parameters which appear in Eq. (5) have been already discussed, except the polarization degree P. In our case, we have $P=0.5\eta(\bar{v}/c)$, where $(\bar{v}/c)=0.67$ is the positron longitudinal polarization at emission averaged over the β^+ spectrum, η is a factor which accounts for the depolarization undergone by positrons during slowing, and the coefficient 0.5 arises from the geometrical efficiency (in fact only those positrons are utilized that are emitted in a hemisphere whose axis coincides with the direction field). Keeping $\eta=1$, Eq. (5) gives for the counting asymmetry the values collected in Table II. In the same table the experimental asymmetries are also reported; their sign shows that the positron helicity is positive, i.e., the positrons are right handed. The magnitude observed for the residual polarization ($\eta=0.74$) turns out to be very high; this means that we must regard as rather ineffective, all those depolarizing phenomena which can act on positrons from the emission to the annihilation. In particular, we refer to depolarization arising from scattering in the source, in the source backing, and in the absorber where positrons are thermalized and positronium forms, and from departure in alignment on behalf of positronium itself before annihilating. Unfortunately, it is not possible at present to explain this result quantitatively on theoretical grounds, owing to the fact that the difficulties encountered in treating depolarization problems increase when one needs to consider the slowing down of positrons down to thermal energy.