Polarization of Positrons and Annihilation in Ferromagnetic Materials*

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The angular correlation of two-quantum radiation from annihilation of positrons in ferromagnetic media has been investigated as a function of the direction of magnetization. The observed changes in the correlations are attributed to the polarization of positrons emitted from an unpolarized source. In the part of the two-quantum correlation associated with large electron momentum, the yield is greater when the polarization of the positrons is parallel to the magnetic field and it is less when the field is reversed. This effect is enhanced when the initial energy, and hence the polarization, of the positrons increases. The qualitative features of the effect were the same in three samples of iron (one containing a large amount of cobalt). No effect was detected for nickel or gadolinium (below the Curie point), and none for any nonferromagnetic metal which was used as a test of the experiment.

INTRODUCTION

HE longitudinal polarization of positrons in beta decay^{1,2} provides a powerful tool not only for understanding the process of beta decay but also for studying various problems in the field of solid state. Page and Obenshain³ have investigated the effect of polarization in the formation of positronium in certain dielectrics in a magnetic field. In a ferromagnetic material² the property of polarization may be used to probe the distribution in momentum and space of the electrons responsible for ferromagnetism. Since our earlier work, we have investigated iron in greater detail and examined a number of other ferromagnetic substances. In addition, the technique has advanced sufficiently to provide a more quantitative connection between the degree of polarization of the positrons and the properties of the ferromagnet.

The aim of the investigation is to study the angular correlation of two-quantum radiation arising from the annihilation of polarized positrons with electrons in a magnetized medium and, upon reversing the direction of magnetization, to detect changes in the correlation which may be attributed to the polarization of the positrons. Since the shape of the correlation curve is related directly to the distribution in momentum of the electrons,^{4,5} a change in a given portion of the curve may be associated with a particular range of electron momenta. If an increase in yield is observed in some part of the curve when, for example, the magnetization is parallel to the direction of polarization of the positrons, this increase may then be attributed to the favorable alignment of the spin of the positrons relative to the spin of electrons⁶ in the specified range of momentum. Since thermalized positrons must remain predominantly at a distance from atomic nuclei, the magnitude of this increase provides information on the spatial distribution of these electrons.

METHOD

The angular correlation of two-quantum radiation was observed in an apparatus having cylindrical symmetry about the two-quantum axis. This would seem to be a natural symmetry for studying two-quantum correlations⁷ and it was ideally suited to the present investigation. The arrangement is shown schematically in Fig. 1.

The small counter (SC), which serves to define the two-quantum axis, was equipped with a collimator having a cylindrical aperture 1 cm in diameter. A few observations were made at a distance of 600 cm so that the aperture subtended at the source an arc of 1.7 milliradians. For most of the measurements the distance was 300 cm corresponding to 3.3 milliradians.

The large counter (LC) itself subtended an angle of 60 milliradians. A typical collimator for this counter is composed of a ring and a plug with tapered sides arranged so as to allow only quanta traveling between θ_1 and θ_2 to reach the detector. The angles are measured from the axis of the detector. A collection of rings and plugs was available so that θ_2 , the outside angle of the collimator, could be varied in steps of two milliradians, and θ_1 , the inside angle, in steps of one milliradian from 0 to 20 milliradians. By combining plugs and rings appropriately, any portion of the angular correlation curve could be singled out for study with crude or fine resolution. For example, a differential angular corre-



FIG. 1. Experimental arrangement.

 $[\]ast$ Work performed under the auspices of the U. S. Atomic Energy Commission.

¹L. A. Page and M. Heinberg, Phys. Rev. 106, 1220 (1957).

 ² S. S. Hanna and R. S. Preston, Phys. Rev. 106, 1363 (1957).
³ L. A. Page and F. E. Obenshain, Bull. Am. Phys. Soc. Ser. II, 260 (1957)

^{2, 260 (1957).} ⁴ DeBenedetti, Gowan, Konneker, and Primakoff, Phys. Rev. 77, 205 (1950). ⁵ D. M. D. M. L. Difference (1956). ⁶ D. D. L. Difference (1956). ⁶ D. D. L. Difference (1956). ⁷ D. D. L. Difference (1956). ⁷ D. D. L. Difference (1956). ⁸ D. D. Difference (1957). ⁸ D. D. Difference (1957). ⁸ D. D. Difference (1957). ⁹ D. Difference

 $^{{}^{5}}$ R. A. Ferrell, Revs. Modern Phys. 28, 308 (1956); S. Berko and F. L. Hereford, Revs. Modern Phys. 28, 299 (1956). These articles provide a review of the extensive literature in the field.

⁶ L. A. Page, Phys. Rev. 106, 394 (1957).

⁷ A similar technique has been used by K. A. Baskova and B. S. Dzhelepov, Izvest. Akad. Nauk S.S.S.R. Ser. Fiz. **20**, 951 (1956).

lation with good resolution could be obtained by using a sequence of zones with $\theta_2 - \theta_1 = 1$ milliradian. On the other hand, an integral angular correlation is measured if only a sequence of inner plugs (or outer rings) is employed.

The housing for the source is shown in Fig. 2. The principal shielding was provided by two parallel slabs of lead. The source of positrons rested in a well in one of the slabs. A small extension of the pole face opposite the source protruded through the other lead slab into the gap between the slabs and carried on its end the sample in which the annihilations occurred. The magnetic field was either parallel or antiparallel to the positron beam and, for convenience of construction, the two-quantum axis was perpendicular to this direction.

The lead slabs provided shielding but not collimation for the counters. In the vicinity of the sample the gap space was 0.125 in. By narrowing this gap to 0.050 in. in the extension of the shielding toward the small

TABLE I. Values of $(N_+-N_-)/N_-$ in percent, for various samples and several thicknesses of absorber. The symbols N_+ and N_- represent the integral yield over angles greater than 8 milliradians, for a field parallel (+) and antiparallel (-) to the direction of motion of the positrons.

	Thickness of aluminum absorber in mils							
	0	4	8	12	16			
	Average energy of positrons in Mev							
Sample	0.33	0.38	0.43	0.47	0.50			
Fe-Co Fe-Co (thin source)	5.4 ± 0.8 4.4 ± 1.2	6.0±1.0	9.4±1.2	11±1.9	11 ± 2.5			
Fe (steel)	4.2 ± 0.8	6.0 ± 1.1						
Fe (rectangular) Fe (Armco) Ni	-03+09	9.0 ± 1.2	8.0 ± 1.4					
Cu Gd (20°C) Gd (~100°C)	0.2 ± 0.9	0.1 1.1.0	0.1 ± 1.2 2.2 ± 3.0 0.0 ± 1.8					
			0.0±1.8					

counter, spurious coincidences were reduced from 5% to 2%. In addition to the basic shielding surrounding the source and the counters, which suppressed undesired coincidences, various auxiliary shields were used, as shown for example at the right in Fig. 2, in order to reduce the noncoincident rate in each counter. A telescope, mounted beyond one of the counters and along the desired axis, was used to align the collimators of both counters with the source of coincident gamma rays.

Throughout the investigation, the source of positrons was Cu⁶⁴ with a half-life of 12.8 hr. The standard source was a copper disk $\frac{3}{16}$ in. in diameter and 0.005 in. thick, irradiated for a period of 24 hours or more in the Argonne research reactor. The source was installed within 2 hours after irradiation and used for a period of 12 to 24 hours. The initial intensity of the source was approximately 50 mC. Relative to a thin source, a rather thick source has the advantage that the increased counting rate is derived from positrons of high-energy and maximum polarization originating in the rear of the source. When, indeed, a source 0.002 in. thick was tried the result indicated, if anything, a smaller average



FIG. 2. Placement of source and sample between the magnet pole pieces and the lead shields.

polarization for those positrons emerging from the source (see Table I). At an early stage in the investigation the backing of the source was changed from lead to Lucite without appreciably altering the results, indicating that backscattering was not producing a serious depolarization in the beam of positrons.

It was estimated that the maximum range of the positrons from a Cu⁶⁴ source, $E_{\rm max} = 660$ kev, was about 0.014 in. in metals such as iron or copper. Accordingly, the samples in which the annihilation occurred were at least 0.015 in. thick. In most cases they were circular, $\frac{3}{16}$ in. in diameter. Since, however, the maximum scattering of annihilation quanta along a diameter of such a sample amounts to about 25%, a rectangular sample $\frac{3}{32}$ in. wide was also used. As expected, the amount of distortion in the angular correlation was apparently reduced and the polarization effect correspondingly enhanced with this sample (Table I).

The magnet had a simple C-shaped yoke with tapered pole pieces as shown in Fig. 2. A magnetomotive force of approximately 20 000 ampere-turns produced an average field of about 15 kilogauss at the surface of the annihilating sample.

Both detectors were conventional sodium iodide counters. Coincidences were detected in a fast-slow circuit having an over-all resolving time of about 10 millimicroseconds and providing energy discrimination in both channels. For almost all observations, both channels were set to accept only pulses in the photopeak of the annihilation radiation, so as to reduce coincidences involving scattered radiation. In all cases at least one of the channels was restricted to 0.5-Mev radiation.

In order to normalize the measurements it was necessary to determine the number of positrons striking the sample during an observation. This was done in two ways. The number was monitored simultaneously by observing the counting rate in the small counter. This method suffers only from the presence of a small but not negligible background of noncoincident radiation. Alternatively, a total two-quantum rate was measured by removing the collimator from the large counter. This procedure depends only on the constancy of the counting equipment during the periods with and without the collimator. Over a long period of time both techniques gave the same results.

TABLE	II.	Saturation	magnetizations	and	Curie	points of
		ferro	magnetic elemen	ts.ª		

		Bohr magneton number		
Element	Curie temp, °K	0°K	300°K	
Fe	1043	2.2	2.1	
Co	1404	1.7	1.7	
Ni	631	0.6	0.55	
\mathbf{Gd}	289	7.1	6.3ь	

^a From American Institute of Physics Handbook (McGraw-Hill Book Company, Inc., New York, 1957). ^b For a temperature of 173°K.

When the magnetic field is turned on, the focusing action of the field produces a marked increase in the number of positrons reaching the sample. This increase in positron flux is accompanied by a decrease in the polarization of positrons reaching the sample, since positrons of low energy, even when traveling at large angles to the direction of the field, are brought to a focus on the sample. In almost every run the amount of focusing was independent of the sense of the magnetic field, as it should be if the source and sample are properly aligned.

In order to discover possible defects in the experimental arrangement, several tests were carried out including the obvious, but important, one of reversing the direction of the positrons relative to the magnet and the detectors. Although all such tests indicated the absence of spurious effects, a null result was obtained for each sequence of runs by replacing the magnetic sample with a nonmagnetic sample of copper.

RESULTS

The principal results fall into two categories: (1) an investigation of various ferromagnetic substances, iron, an iron-cobalt alloy, nickel, and gadolinium, and (2) a determination of the degree of polarization as a function of positron energy. All the results in Table I were obtained under essentially identical conditions except for the differences noted. The magnitude of the magnetic field was the same in all cases except for the negligible perturbations introduced by the different samples themselves, which were alike in size and shape. Other factors, such as the shape and thickness of the source, were kept virtually unchanged.

Since the differential angular correlations (see following material) did not reveal significant fine structure, the measurements in Table I are of the integral type obtained with one of the cylindrical plugs in front of the large counter. The eclipsed portion of the counter sub-

TABLE III. Spectrochemical analysis of samples in percent. The X indicates the major constituent. Three dots signify an undetectable amount, less than 0.01%.

Sample	Fe	Co	Ni	Cu	Mg	Mn	Si	С	Gd
Fe (steel)	X		0.02			0.1	0.01	0.16	
Fe (Armco)	X		0.02					0.02	
Fe-Co	63.2	37.7	0.1						• • •
Ni	0.05	0.1	X	0.01	0.01	0.05	0.01	0.14	
Cu				X				0.01	• • •
Gd	0.3								X

tended a half angle of 8 milliradians at the source, an angle chosen to obtain a good polarization effect as well as a substantial counting rate. The numbers in Table I are measured values of $(N_+ - N_-)/N_-$, where N represents the total integrated yield of coincidences over angles greater than 8 milliradians. The + and - denote, respectively, measurements with the magnetic field parallel and antiparallel to the positron beam. The average energy of the positrons was varied by inserting aluminum foils of various thicknesses between source and sample. The estimated average energy is recorded in the table.

The pertinent magnetic properties of the ferromagnetic elements are listed in Table II. For cobalt, iron,



FIG. 3. Integral angular correlations for annihilation in iron for a magnetic field parallel (+) and antiparallel (-) to the direction of motion of the positrons. Measurements are normalized to the total coincident yield.

and nickel the Curie temperatures are well above room temperature so that the magnetization is essentially complete without cooling. For gadolinium, however, it was necessary to cool the sample. This was done by attaching the small iron piece which holds the sample to a copper bar immersed in liquid nitrogen. It was further necessary to enclose the region between the pole faces of the magnet in a dry atmosphere so as to prevent the formation of ice on the sample. The composition of each sample, as determined by spectro-chemical analysis, is given in Table III.

The angular correlations which have been measured so far are reproduced in Figs. 3–5. More detailed measurements are planned. The data, however, give the general trend of the correlations and serve as a guide for future work. The curves are presented as they were obtained experimentally. For the integral correlations, therefore, the intensity at the angle θ (the half-angle of the plug of the collimator) represents the total yield of coincidences over all angles greater than θ . The solid angle for each point is equal to $2\pi(1+\cos\theta)$, a slowly varying function over the region studied. Since the measurements are normalized to the total coincident yield, the intensity at $\theta=0$ is equal to unity. For the differential correlation the solid angle varies as

$$2\pi(\cos\theta_1-\cos\theta_2)\cong 2\pi(\theta_2-\theta_1)\sin\left[\frac{1}{2}(\theta_2+\theta_1)\right],$$

where $\theta_2 - \theta_1$ is the constant angular aperture, equal to 2 milliradians in this case, and $(\theta_2 + \theta_1)/2$ is the mean angle of observation. The decrease in the solid angle accounts for the fall in the correlation function as θ_1 approaches zero. The points have been plotted against θ_1 , the inner angle of the collimator opening.

It is apparent from these correlations that no prominent structure is introduced by the magnetic field and the difference between the curves for positive and negative field varies smoothly from point to point.

TABLE IV. Values of $(N_+' - N_-')/N_-'$ in percent, for various samples. The symbols N_+' and N_-' represent the integral yield through an aperture, i.e., over angles less than θ , where θ is the half-angle listed, for field parallel (+) and antiparallel (-) to the direction of motion of the positrons.

Sample	Half-angle milliradians	Al absorber mils	$(N_{+}' - N_{-}')/N_{-}'$
Polvethylene	2	8	$+4.0\pm1.4$
$Gd(-100^{\circ}C)$	2	8	-1.3 ± 2.2
Fe	6	4	-3.2 ± 1.3

Inside about 8 milliradians the differential yield is greater when the field is opposite the direction of the positrons; outside this angle the effect is reversed.

Data taken with a ring collimator without the inner plug are presented in Table IV. In these observations the central cone of the angular distribution is isolated for study. The negative value for iron serves merely to confirm the positive results of Table I obtained with the central cone eclipsed. The observation on gadolinium was made with a narrow aperture in order to investigate the very center of the two-quantum distribution, since the earlier investigation (Table I) had shown very little effect outside, and hence inside, a larger region. However, no effect was obtained.

The measurement on polyethylene was undertaken to provide a link between the measurements of Page and Obenshain³ and the present observations. The use of a narrow aperture makes the technique analogous to that of Page and co-workers.^{1,3} Page and Heinberg observed the effect of a magnetic field on the twoquantum annihilation of positronium formed in argon gas, by the positrons from Na²², and obtained a result consistent with a negative value for our ratio $(N_+'-N_-')/N_-'$. Page and Obenshain state that their results using polyethylene and fused quartz, instead of



FIG. 4. Differential angular correlations for annihilation in iron for a magnetic field parallel (+) and antiparallel (-) to the direction of motion of the positrons. The angular aperture $\Delta\theta=2$ milliradians.

argon, have just the opposite dependence on the sign of the magnetic field. Hence the positive sign obtained here for polyethylene serves to establish the mutual consistency of the two experiments and the measurement provides an interesting comparison of the two different experimental techniques.[†]



FIG. 5. Integral angular correlations for annihilation in the iron-cobalt sample for a magnetic field parallel (+) and antiparallel (-) to the direction of motion of the positrons.

 \dagger Note added in proof.—Dr. Page has kindly informed us that he has made observations on an iron sample in his apparatus, and has also confirmed the consistency of the results.

DISCUSSION

As discussed in our earlier communication,² it is natural to attribute the changes observed in the angular correlation to the polarization of the positrons, which in turn is a consequence of the fundamental modification in the concept of parity for weak interactions.⁸ If the polarization of the positrons is parallel to their momentum, the sense of the observed effect is such that when the magnetization is parallel (electron spin antiparallel) to the positron spin the two-quantum yield is enhanced in the region of the angular correlation associated with high momentum.

Table II shows that, below their Curie points, iron, cobalt, nickel, and gadolinium are ferromagnetic in varying degrees. The different results obtained for these elements and their alloys reflect not only the gross differences in their ferromagnetic properties, but also more sensitively the variations in the spatial and momentum distributions of electrons which are polarized in a magnetic field. It is seen in Table I that the iron samples show the greatest effect. The less strongly ferromagnetic nickel and the nonferromagnetic materials, gadolinium at room temperature and copper, show little or no effect. It is not immediately apparent whether the unexpected result for nickel should be attributed to a significant difference in the momentum distribution of the magnetic electrons in nickel, as compared to iron and cobalt, or to a relative scarcity of these electrons in the region penetrated by the positron waves in nickel. Gadolinium cooled to -100 °C is strongly ferromagnetic but no effect is observed either in the wings of the distribution or in its central peak. In this case the polarized electrons are presumably quite effectively shielded from the positrons.

If the degree of polarization at the time of emission of a positron is equal to v/c,⁹ an approximate value of the polarization at the time of annihilation is obtained by taking account of the following factors: (1) the shape of the beta spectrum, (2) the angle of acceptance for the positrons in the presence of the magnetic field, and (3) the absorption of positrons of low energy in the



FIG. 6. Sketch illustrating the region of momentum space eclipsed when the cylindrical absorber is used as a collimator. The quantity $4\pi p (p^2 - p_c^2)^{\frac{1}{2}} dp$ is the volume of that part of the spherical shell which is outside the cylinder of eclipse.

source and the absorbing foil. The effects of backscattering and depolarization in slowing down are not included in the calculation. With the maximum thickness of aluminum absorber, the average polarization is $P_p = \langle v/c \rangle = 0.88$ for positrons emerging from the absorber and traveling parallel to the magnetic field. It is estimated that the polarization is lowered to 0.70 when all the positrons focused on the sample are considered.

It is convenient to define for the electrons participating in annihilation a polarization expressed (as for the positrons) in the usual way: $P_e = (n_+ - n_-)/(n_+ + n_-)$, where n_+ and n_- stand, respectively, for the numbers of electrons with spin parallel and antiparallel to the field. The ratio $R = N_+/N_-$ is then given by the usual formula,

$$R = (1 + P_e P_p) / (1 - P_e P_p).$$

After rearrangement we obtain

$$P_e = \frac{1}{P_p} \left(\frac{R-1}{R+1} \right).$$

Correction for the scattering of gamma rays in the iron-cobalt sample changes the result from the observed R=1.11 to approximately R=1.12. With $P_p=0.70$, we then find $P_e=0.08\pm0.03$. For the other iron samples a similar value would be obtained.¹⁰

This value for the effective polarization applies, of course, only to those electrons in the sample which are subject to annihilation and at the same time satisfy a condition on their momenta. If p is the momentum and $4\pi p^2 \eta(p)$ the momentum distribution for the electrons subject to annihilation, then the distribution for electrons whose annihilation can be detected in a measurement of the integral type is (see Fig. 6)

$$4\pi p (p^2 - p_c^2)^{\frac{1}{2}} \eta(p)$$

where p_c is the smallest momentum allowed by the half-angle of the plug of the collimator. The total number of contributing electrons is, therefore,

$$4\pi \int_{p_c}^{\infty} p(p^2 - p_c^2)^{\frac{1}{2}} \eta(p) dp.$$

For an angle of 8 milliradians, p_c corresponds to an electron energy of 16 ev.

It is clearly demonstrated in Table I that for the iron-cobalt alloy the measured value of $(N_+ - N_-)/N_-$ increases as the average energy of the positrons increases. Only a small amount of this enhancement can be attributed to the change in v/c, which varies only from about 0.80 to 0.88. Most of the increase is due to the improved directionality and correspondingly greater polarization in the emergent beam of positrons which results from the reduction in scattering for very energetic positrons.

⁸ T. D. Lee and C. N. Yang, Phys. Rev. 104, 254 (1956); 105, 1671 (1957).

⁹ Jackson, Treiman, and Wyld, Phys. Rev. 106, 517 (1957).

¹⁰ It is interesting to note that the polarization of *all* electron in iron at magnetic saturation is (14-12)/26=0.08.