Annihilation of Positrons in Flight*†

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The weak continuous spectrum of hard γ rays resulting from the stopping of positrons in various materials has been investigated with a NaI(Tl) scintillation spectrometer. Measurements were made chiefly in the γ-ray energy interval from 0.511 Mev to 1.3 Mev, for the continuous β spectra of Ne^{19} and A^{35} (maximum positron energies of 2.2 Mev and 4.4 Mev, respectively). The experimental geometry was chosen to ensure averaging of the annihilation spectrum over all angles. Theoretical spectra were calculated for Lucite, brass, and lead as stopping materials. Comparison with experiment on an absolute basis was made possible by an experimental determination of the spectrometer efficiency as a function of γ-ray energy. The calculations included bremsstrahlung and single-quantum annihilation as well as two-quantum annihilation. In the case of Ne^{19} positrons annihilating in brass, the data are in satisfactory agreement with the calculations. However, the calculated spectra show a conspicuously stronger dependence on the atomic number of the stopping material than do the observed spectra. The experimental spectra from Ne^{19} positrons stopping in Lucite and brass are indistinguishable, while the calculated spectra show a brass to Lucite ratio of 1.3. In addition, the calculated intensities tend to be lower than the observed ones, particularly for A^{35} positrons and for stopping materials of low atomic number.

I. INTRODUCTION

IT has been demonstrated several times that positrons annihilate primarily after they reach the end of their paths of ionization and have only thermal energies. However, there is a small probability that annihilation resulting in two quanta will occur before a positron loses its kinetic energy.[1,2] A large number of experimenters have reported evidence for this annihilation process.[3–19] Their results gave little information, beyond confirming that positrons do annihilate in flight, until the work of Colgate and Gilbert[16] with high-energy positrons and that of Deutsch *et al.*[14,18] with low-energy positrons stopping in anthracene. The present authors were led to the study of hard annihilation radiation by the appearance of a hard γ-ray continuum[20] in the radiation accompanying the β decay of C^{10}, which has subsequently been found in the decays of the positron emitters C^{11}, O^{15}, Ne^{19}, and A^{35}.

II. THEORETICAL CALCULATIONS

Using Dirac's expression[1] for the probability of transition to a negative energy state, Bethe[2] obtained the cross section for two-quantum annihilation of a positron in flight with a free electron at rest:

$$d\Phi=\frac{\pi r_0^2}{\epsilon^2-1}\left[\frac{k}{k'}+\frac{k'}{k}+2\frac{\epsilon+1}{kk'}-\left(\frac{\epsilon+1}{kk'}\right)^2\right]dk. \quad (1)$$

The classical electron radius is $r_0=e^2/mc^2$; the energy of the positron, including rest energy, is ϵmc^2; and the energies of the two quanta are kmc^2 and $k'mc^2$, where $k+k'=\epsilon+1$. The conservation laws require the photon energies to lie between the extreme values

$$k=\tfrac{1}{2}(\epsilon+1)\pm\tfrac{1}{2}(\epsilon^2-1)^{\frac{1}{2}}, \quad (2)$$

corresponding to emission in the forward and backward directions.

The probability that a positron annihilates with emission of a photon in the range dk while traversing a path length dx is then

$$NZd\Phi dx=NZd\Phi(d\epsilon/dx)^{-1}d\epsilon, \quad (3)$$

N being the number of atoms per unit volume with Z electrons per atom. The average rate of collision loss by positrons is

$$-\frac{d\epsilon}{dx}=NZ\frac{2\pi r_0^2}{\beta^2}\left\{\ln\frac{2T^2(\epsilon+1)}{I^2}-\frac{\beta^2}{12}\left[23+\frac{14}{\epsilon+1}+\frac{10}{(\epsilon+1)^2}+\frac{4}{(\epsilon+1)^3}\right]\right\}, \quad (4)$$

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[17] C. E. Violet, thesis, University of California Radiation Laboratory Report UCRL-2163, 1953 (unpublished).
[18] H. W. Kendall and M. Deutsch, Phys. Rev. **93**, 932 (1954). We are indebted to these authors for allowing us to see a fuller (unpublished) account of this work.
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[20] R. Sherr and J. B. Gerhart, Phys. Rev. **91**, 909 (1953).

where $\beta^2=(\epsilon^2-1)\epsilon^{-2}$ is the square of the velocity in units of c, $T=(\epsilon-1)mc^2$ is the kinetic energy, and I is an average ionization potential for an atom of the stopping material. This rate of collision loss is obtained[21] from the Bhabha cross section for positron-electron scattering; it differs by about 5 percent at 100 kev, and by less at higher energies, from the Bethe formula for collision loss by negative electrons.

If every positron started out with the same initial energy ϵ_1, we could obtain the number of annihilation photons in the range dk per positron by integrating (3) over positron energy from ϵ_1 down to the lowest value at which the positron still has sufficient energy to produce a photon in the range dk. This minimum value is found from (2) to be

$$\epsilon_m=k-(k-1)/(2k-1). \tag{5}$$

The resulting spectrum would not be quite correct, since it has been implicitly assumed that each positron passes through the range $d\epsilon$ in the course of slowing down. This is surely not the case for those positrons that annihilate at energies higher than the range in question, but they are a small fraction of the total number of positrons and can be neglected to a fairly good approximation. The errors introduced by disregarding this attenuation by annihilation will be estimated later.

If the positron source is a β emitter, the annihilation spectrum from positrons of a given initial energy can be determined in the manner just described, and the results can then be averaged over the β spectrum of initial energies. For each value of photon energy two numerical integrations are required by this procedure, one over the energy of the stopping positron and the second over initial energies. It is therefore easier to interchange the order of integration and to determine first the fraction of positrons from the β spectrum that pass through a given energy range $d\epsilon$ in the course of slowing down. The probability Q of passing through the interval ϵ to $\epsilon+d\epsilon$ is simply the probability of having an initial energy above this range (attenuation by annihilation again being neglected)

$$Q(\epsilon,\epsilon_0)=\int_{\epsilon}^{\epsilon_0} q(\epsilon,\epsilon_0)d\epsilon, \tag{6}$$

where $q(\epsilon,\epsilon_0)$ is an allowed Fermi spectrum, normalized to unity, with end point ϵ_0. (All the positrons used in the present experiments came from allowed β transitions, and the Coulomb correction to the spectrum was found to be negligible for present purposes in the source with largest atomic number, A^{35}.) The probability (3) that a positron gives rise to an annihilation quantum k, assuming that it reaches the energy interval $d\epsilon$ in the course of slowing down, is now multiplied by the probability Q that it does in fact reach this interval, and the product is integrated from ϵ_0 to ϵ_m, i.e., over the range of annihilation energies compatible with k. Thus the number of photons in the interval dk per positron of the β spectrum is found by a single numerical integration for each value of k, the integration (6) being independent of k:

$$n(k,\epsilon_0,Z)dk=dk\int_{\epsilon_m}^{\epsilon_0} NZ(d\Phi/dk)\times(-d\epsilon/dx)^{-1}Q(\epsilon,\epsilon_0)d\epsilon. \tag{7}$$

Since the average energy loss (4) is proportional to the number NZ of electrons per unit volume, the annihilation spectrum (7) depends on the stopping material only through the logarithm of the ionization potential. This dependence is sufficient to make the hard annihilation spectrum from positrons stopping in lead 15 to 20 percent higher than in brass.

Figures 1 through 4 show $n(k,\epsilon_0,Z)$ for Ne^{19} and A^{35} positrons annihilating in brass and lead. Note that the

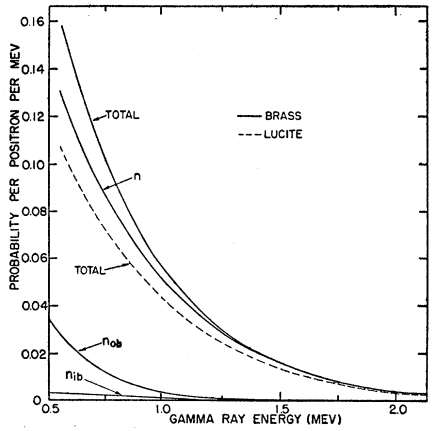

FIG. 1. Theoretical γ-ray spectrum for 2.2-Mev Ne^{19} positrons annihilating in brass and in Lucite.

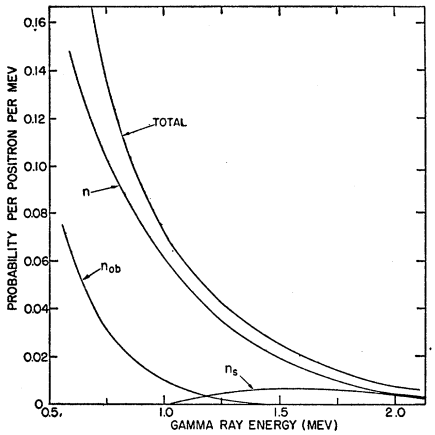

FIG. 2. Theoretical γ-ray spectrum for 2.2-Mev Ne^{19} positrons annihilating in lead. n_{ib} (omitted here) is the same as in Fig. 1.

[21] F. Rohrlich and B. C. Carlson, Phys. Rev. 93, 38 (1954).

curves shown in the figures are plotted with Mev as the energy unit rather than mc^2; the change in units introduces only a constant numerical factor.

In addition to two-quantum annihilation there are three other sources of hard γ radiation for which corrections to the calculated spectrum (7) must be made: single-quantum annihilation, bremsstrahlung produced in the stopping material, and inner bremsstrahlung produced in the source itself.

Bethe[2] gives the cross section per K electron for single-quantum annihilation:

$$\Phi_s(\epsilon,Z)=\alpha^4Z^5 2\pi r_0^2(\epsilon+1)^{-2}(\epsilon^2-1)^{-1} \times[(\epsilon^2-1)^{\frac{1}{2}}(\epsilon^2+\tfrac{2}{3}\epsilon+\tfrac{4}{3})-(\epsilon+2)\cosh^{-1}\epsilon], \quad (8)$$

where $\alpha=1/137$ and ϵmc^2 is the total positron energy, including rest energy. The energy of the emitted photon

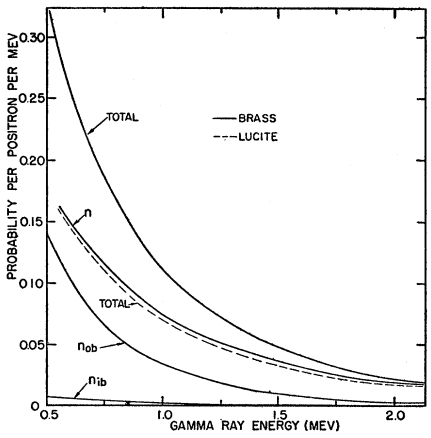

FIG. 3. Theoretical γ-ray spectrum for 4.4-Mev A^{35} positrons annihilating in brass and in Lucite.

is given to a sufficient approximation by $k=\epsilon+1$, without taking into account the binding energy of the K electron or the loss of kinetic energy by the positron in penetrating the Coulomb field. The probability that a positron from the β spectrum will annihilate by this process in the range $d\epsilon$ is

$$N2\Phi_s(\epsilon,Z)(d\epsilon/dx)^{-1}Q(\epsilon,\epsilon_0)d\epsilon, \quad (9)$$

and the resulting photon spectrum $n_s(k,\epsilon_0,Z)dk$ is equal to the probability (9) evaluated at $\epsilon=k-1$, with $d\epsilon$ replaced by dk. The factor 2 in (9) represents the two K electrons per atom; contributions from electrons in higher shells are neglected. Since (9) depends on the stopping material approximately as the fourth power of Z, single-quantum annihilation is negligible in Lucite and brass, although it makes an appreciable contribution in lead.

The cross section for bremsstrahlung, including screening corrections, was obtained in Born approximation by Bethe and Heitler,[22] and their results have been

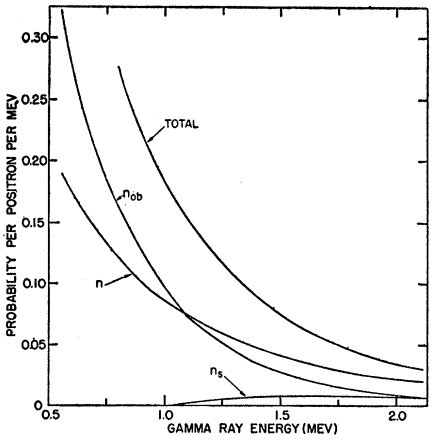

FIG. 4. Theoretical γ-ray spectrum for 4.4-Mev A^{35} positrons annihilating in lead. n_{ib} (omitted here) is the same as in Fig. 3.

plotted for lead in the energy range of a few Mev by Ivanov *et al.*[23] In the present calculations a linear interpolation between the end points of the Bethe-Heitler intensity distribution has been used; the cross section for emission of a photon in the range dk has been taken to be

$$d\Phi_{ob}=k^{-1}dk\alpha Z^2r_0^2 20[1-k/(\epsilon-1)]. \quad (10)$$

This vanishes at the short-wave limit, as does the Born approximation. Although Jaeger[22] has shown that the exact cross section for negative electrons is different from zero at the short-wave limit, the exact cross section for positrons would be expected to vanish there, as in the nonrelativistic theory. The spectrum of radiated photons per positron is found from (10) by numerical integration:

$$n_{0b}(k,\epsilon_0,Z)dk=dk\int_{k+1}^{\epsilon_0}N(d\Phi_{0b}/dk) \times(-d\epsilon/dx)^{-1}Q(\epsilon,\epsilon_0)d\epsilon. \quad (11)$$

It is approximately proportional to the Z of the stopping material.

Finally, the contribution of inner bremsstrahlung has been obtained from the probability $d\Phi_{ib}$, given by Knipp and Uhlenbeck,[24] for emission of a photon in the range dk and an electron of energy $(\epsilon-k)mc^2$ in an allowed β transition. The resulting photon spectrum, which is of course independent of the stopping material, is obtained by an integration over the differential Fermi spectrum

$$n_{ib}(k,\epsilon_0)dk=dk\int_{k+1}^{\epsilon_0}(d\Phi_{ib}/dk)q(\epsilon,\epsilon_0)d\epsilon. \quad (12)$$

[22] H. Bethe and W. Heitler, Proc. Roy. Soc. (London) **A146**, 83 (1934). See also W. Heitler, *The Quantum Theory of Radiation* (Oxford University Press, London, 1944). The accuracy of the Born approximation is discussed by J. C. Jaeger, Nature **140**, 108 (1937).

[23] Ivanov, Walter, Sinelnikov, Taranov, and Abramovich, J. Phys. (U.S.S.R.) **4**, 319 (1941).

[24] J. K. Knipp and G. E. Uhlenbeck, Physica **3**, 425 (1936). See also F. Bloch, Phys. Rev. **50**, 272 (1936).

The correction for inner bremsstrahlung proves to be a very small one (see Figs. 1–4).

Conversion of the calculated photon spectra (7), (9), (11), and (12) to pulse-height distributions will be discussed in Sec. V.

III. EXPERIMENTAL PROCEDURE

The positron activities studied were produced by bombardment of powders with 18-Mev protons from the internal beam of the Princeton cyclotron.[25] By continuously sweeping helium through the probe, the desired activities were carried off in gaseous form to a suitable gas chamber where their decays could be observed at fairly constant counting rates with a NaI(Tl) scintillation spectrometer. A trap containing ascarite and $CaCl_2$ was placed in the gas line between the probe and the observation chamber to remove possible extraneous activities in the form of CO_2 or water vapor.

The activities studied were primarily Ne^{19} and A^{35} (end-point energies 2.2 and 4.4 Mev, respectively). Ne^{19} was produced through the reaction $F^{19}(p,n)Ne^{19}$ by bombarding PbF_2 powder. $PbCl_2$ powder was bombarded to give A^{35} through the reaction $Cl^{35}(p,n)A^{35}$. In both cases the activities were identified by half-life measurements, by determination of the positron energies in absorption measurements, and by the absence of nuclear γ radiation.

C^{10} and C^{11} (end-point energies 2.1 and 0.97 Mev, respectively) were produced simultaneously by bombarding B^{10} powder. C^{10} resulted from the $B^{10}(p,n)C^{10}$ reaction; C^{11} from the $B^{11}(p,n)C^{11}$ reaction on B^{11} impurity in the target. Both activities were carried off as CO_2, the ascarite-$CaCl_2$ trap being removed from the gas line in this case and the inspection chamber charged with ascarite to absorb the activity. The C^{10} spectrum could be observed directly inasmuch as the C^{11} contributed only a small fraction of the total activity. To observe the 20-minute C^{11} activity, gas was allowed to flow through the gas chamber until sufficient activity had been trapped. The gas flow was then interrupted and the 20-second C^{10} activity allowed to decay through approximately five half-lives before the residual C^{11} activity was observed.

O^{15} (end-point energy 1.7 Mev) was produced by a $F^{19}(p,\alpha n)O^{15}$ reaction accompanying the $F^{19}(p,n)Ne^{19}$ reaction. The O^{15} activity was carried off as water vapor and ordinarily was stopped in the ascarite-$CaCl_2$ trap. To observe it the trap was removed and the gas chamber charged with $CaCl_2$. The gas flow was interrupted after the O^{15} activity had been allowed to collect in the $CaCl_2$ and the Ne^{19} activity was allowed to decay before the longer-lived O^{15} was observed.

The gas chambers used to observe the various activities had walls of sufficient thickness to stop all positrons. Hence, the effective source of annihilation radiation was the walls of the gas chamber. The original chamber was a brass cylinder (interior dimensions: 4.4-cm diameter by 5.2-cm, 0.33-cm walls) which was placed about 2 cm above the NaI(Tl) crystal (Fig. 5). This chamber could be filled with ascarite or $CaCl_2$ to study the C^{10}, C^{11}, and O^{15} activities, and was lined with lead foil to study annihilation in lead. A similar chamber of Lucite (interior dimensions: 4.1-cm diameter by 6.4-cm 1.2-cm walls) was used to observe annihilation in Lucite.

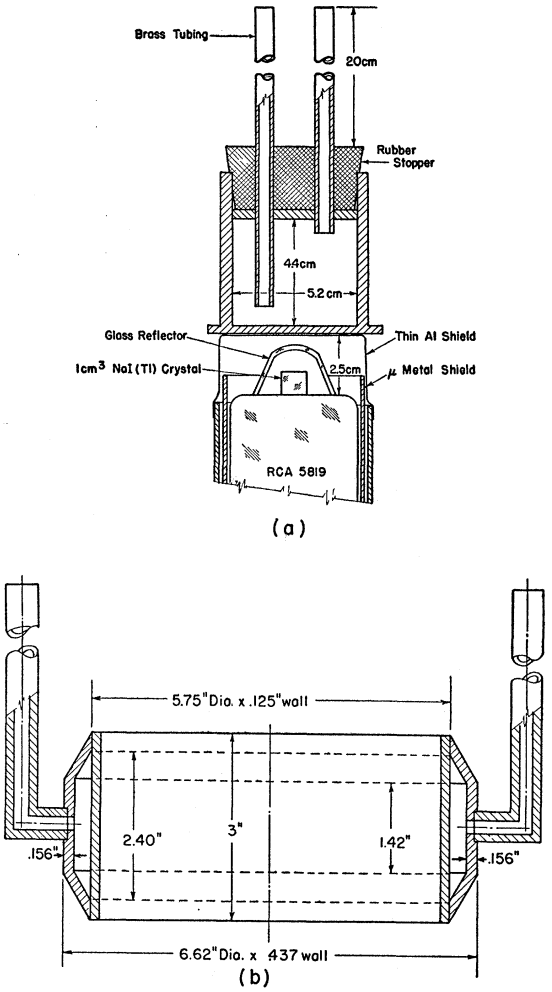

FIG. 5. (a) Cross section of cylindrical brass gas chamber and crystal mounting. (b) Cross section of toroidal brass gas chamber.

The calculations previously outlined assumed that the radiation is emitted isotropically. In the gas chamber just described, positrons enter the walls of the chamber at random and annihilate close to the point of entry to the wall, since the positron range is small. Therefore, we have in effect an extended source for the γ radiation, and the solid angle subtended by the γ-ray detector varies considerably over the chamber. However, if radiation coming from each point of the wall is isotropic, its contribution to the total γ-ray spectrum may vary

[25] Sherr, Muether, and White, Phys. Rev. 75, 282 (1949).

in magnitude but the relative amounts of 0.511-Mev radiation and of hard radiation will be independent of position. Since we use the relative intensities to compare the experimental and theoretical results, the extended source introduces no complications. If each point in the extended source is not isotropic, the resulting spectrum is difficult to interpret. The condition for isotropy will be satisfied if the positrons lose their sense of direction before annihilating. In order to check this point, we repeated the measurements using a brass gas chamber in the form of a hollow toroid with the detector at the center (see Fig. 5). The dimensions were chosen so that the variation of the solid angle of the detector over the toroidal chamber wall was small (less than 10 percent). Hence, no portion of the chamber wall was favored over another as a source of radiation. The chamber was also designed to minimize the possibility of excessive scattering and degradation of the γ rays which reached the crystal. Under these conditions the γ radiation will be isotropic regardless of whether the positrons maintain their original direction or not. It was possible to line a similar chamber with lead foil to study the annihilation spectrum for lead.

With both the cylindrical and the toroidal chambers, less than 5 percent of the radiation reaching the detector originated in the brass pipes leading into the chamber. In Ne^{19} and A^{35} no significant difference was found between the spectra taken with the cylindrical chamber and those taken with the toroidal chamber which could be attributed to geometrical effects. (A small difference caused by unequal γ-ray absorption in the walls of the different chambers was observed.) Consequently, it was assumed that the C^{10}, C^{11}, and O^{15} spectra taken only with the cylindrical brass chamber are free from geometrical effects.

Pulses from the NaI(Tl) crystal, a 1.0-cm cube mounted on an RCA 5819 photomultiplier, were fed to a preamplifier and thence to a Los Alamos model 501 amplifier and recorded on a 10-channel pulse-height analyzer. Both 2- and 5-volt channels were available for inspection of the 0.511-Mev annihilation radiation photopeak and the continuous distribution of hard radiation. A single-channel differential pulse-height discriminator and scalar simultaneously recorded the pulses falling in the 0.511-Mev photopeak and served as a monitor of the effective source strength.

The well-known γ rays of Na^{22}, Sc^{46}, Co^{60}, Zn^{65}, Cs^{134}, Cs^{137}, and Hg^{205} were used to calibrate the pulse-height voltage scale on an absolute basis. Counting rates were kept low enough to prevent pileup and accidental coincidences. Background was less than 5 percent for points at pulse heights corresponding to 1.1 Mev and considerably less for smaller pulse heights.

IV. EXPERIMENTAL RESULTS

In making each run, the 10-channel analyzer was first set to count the 0.511-Mev photopeak pulses in 2-volt channels for approximately 200 seconds. Immediately afterward the hard annihilation radiation was counted with 5-volt channels, the setting of the single channel monitor remaining unchanged, for periods varying from 500 to 2000 seconds. The data obtained in this way were divided by the appropriate channel width and by the simultaneous monitor count. The result is a spectrum independent of both source strength and running time. Separate spectra taken of the same positron source were plotted on an energy calibrated pulse height scale and normalized to the same (arbitrary) 0.511-Mev photopeak area. In this way separate runs were plotted together in a manner eliminating small differences caused by slightly differing monitor settings and amplifier gains. All the data for various sources and stopping materials were normalized finally to the same photopeak area.

The spectra for annihilation in brass were not corrected for absorption of the γ rays in the stopping material. Because of the method of normalization used, the effect of this absorption is to raise the observed hard annihilation spectrum above the true spectrum, but by less than 5 percent. The data for annihilation of positrons in lead and Lucite have been subjected to a correction for the difference in absorption between lead and brass, and between Lucite and brass, respectively. The correction was determined experimentally by inserting a lead absorber of the same thickness as the lead chamber lining between the brass chamber and the detector, and observing the attenuation produced in the annihilation spectrum. A similar procedure was used to determine the correction to the Lucite spectrum.

Figure 6 contains the experimentally observed spectra for the positrons of A^{35} and Ne^{19} annihilating in brass. Both the A^{35} and the Ne^{19} spectra are averages of the two runs made with the toroidal gas chamber which

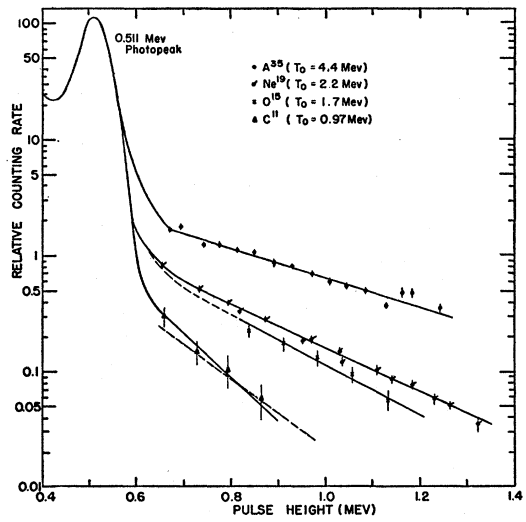

FIG. 6. Observed NaI(Tl) pulse-height distributions for annihilation radiation from positrons stopped in brass. T_0 is the end-point kinetic energy of the positron spectrum. The dashed line passing through the C^{11} points is a calculated spectrum for C^{11}.

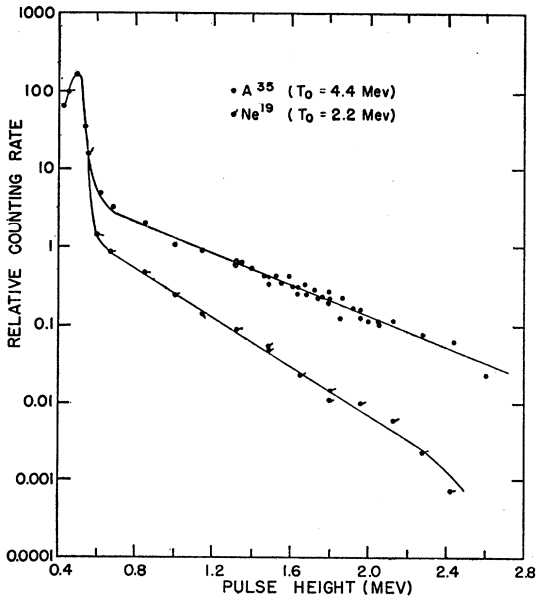

FIG. 7. Observed pulse-height distributions for annihilation of Ne^{19} and A^{35} positrons in brass, showing the absence of high-energy nuclear γ rays. The energy scale shown here is not known to the accuracy of that in Fig. 6. T_0 is the end-point kinetic energy of the positron spectrum.

are freest from instrumental and statistical uncertainties. The A^{35} and Ne^{19} spectra were observed out to higher energies with both 2-volt and 5-volt channels to make certain there were no nuclear γ rays present. None was found, the slopes shown in Fig. 6 persisting out to 2.4 Mev beyond which the counting rates decreased more rapidly until submerged in background counts (see Fig. 7). Also shown in Fig. 6 are the spectra for C^{11} and O^{15} positrons stopping in NaOH and asbestos (Ascarite), and $CaCl_2$, respectively. Although these materials have a lower effective Z than brass, we include these spectra here since brass and Lucite gave the same spectra for Ne^{19}. The data for C^{11} and O^{15} are for single runs. Since the experimental spectrum for C^{11} contains statistical uncertainties too large to distinguish the Z of the stopping material, the Z of brass was used in calculating the spectrum. The theoretical spectrum falls within the limits of the data, as shown in Fig. 6.

The accuracy of the experimental results is indicated by the statistical flags on the individual points. Drifting of the channel widths of the multichannel discriminator introduces a scattering of adjacent points but should not affect the over-all trend of the spectrum. The effect of channel drift is particularly noticeable in the A^{35} spectrum.

The hard annihilation spectrum resulting from the 2.1-Mev positron decay of C^{10} (positrons stopped in ascarite) has been reported previously,[20] and was found to be the same, within experimental error, as the spectrum in brass for the 2.2-Mev positron decay of Ne^{19}.

The data showing the Z dependence of the annihilation spectrum for a single nuclear positron source (Figs. 8 and 9) were taken successively so that the lead and Lucite spectra could be compared with the concurrently observed brass spectra to a better accuracy than had the spectra so compared been observed at different times with different instrumental conditions. The data for annihilation in brass of Figs. 8 and 9 are not the same as shown in Fig. 6.

V. PROCEDURE FOR COMPARISON OF THEORY AND EXPERIMENT

In the present experiments, the annihilation radiation was observed with a NaI(Tl) γ-ray spectrometer; consequently, a calculated γ-ray spectrum, such as that given by Eq. (7), must be converted to a pulse-height distribution before the theoretical calculation can be compared with experiment. Since a large part of the γ-ray spectrum contributes to the counting rate observed at a given pulse height with a NaI(Tl) spectrometer, (7) must be multiplied by a weighting function $f(k,V)$ and integrated over all values of k. In its dependence on pulse height, $f(k,V)dV$ (Fig. 10) gives the probability that a γ ray of energy kmc^2 will produce a pulse in the NaI(Tl) spectrometer of pulse height V (Vmc^2 is the energy of a γ ray whose photopeak maximum falls at V). Relative values of $f(k,V)$ were determined by interpolation between NaI(Tl) spectra for the well-known Au^{198}, Nb^{95}, Zn^{65}, and Na^{22} γ rays as observed with the spectrometer used in this experiment. An experimentally determined[20] photopeak efficiency curve for the spectrometer was used to normalize the spectra to equal source strengths. The functions $f(k,V)$ obtained are accurate to about 5

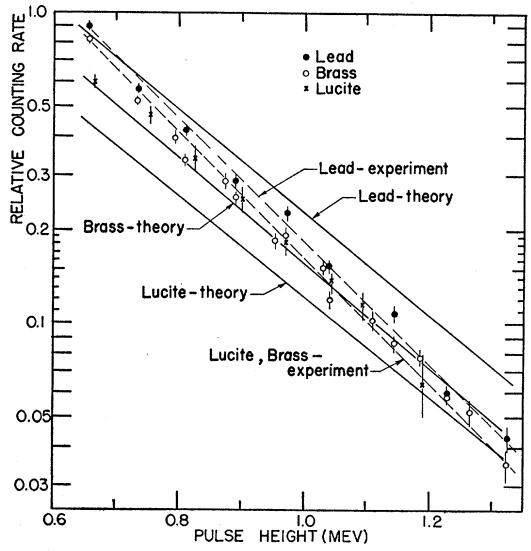

FIG. 8. Comparison of theoretical pulse-height distributions and data for 2.2-Mev Ne^{19} positrons annihilating in lead, brass, and Lucite.

percent. The final calculated spectrum is then given by

$$\mathfrak{N}(V,\epsilon_0,Z)dV = dV\int f(k,V)n(k,\epsilon_0,Z)dk. \quad (13)$$

$\mathfrak{N}(V,\epsilon_0,Z)$ is the number of pulses per unit pulse-height interval per positron in the NaI(Tl) spectrum of the annihilation radiation from nuclear positrons of end point energy ϵ_0 annihilating in material of atomic number Z (see Fig. 10b).

In comparing the calculated distribution (13) with an observed distribution, in which the number of counts per pulse-height interval per second depends on the positron source strength and position, one can avoid a determination of absolute efficiency by using the 0.511-Mev photopeak from annihilation at rest. Since the probability of annihilation in flight is small, there are very nearly two quanta of 0.511 Mev per positron of the β spectrum, and the total spectrum of annihilation quanta per positron is therefore $n(k,\epsilon_0,Z)+2\delta(k-1)$. The second term is a delta function representation of the spectrum of quanta from annihilation at rest. Correspondingly, $\mathfrak{N}(V,\epsilon_0,Z)+2f(1,V)$ is the total pulse-height distribution, whose 0.511-Mev photopeak comes almost entirely from the second term. If this distribution is now multiplied by a constant factor to make the photopeak area of $2f(1,V)$ equal to the photopeak area of the observed distribution, one obtains a direct comparison of the calculated and observed distributions from annihilation in flight. Since all the calibration curves $f(k,V)$ are determined for the same (though arbitrary) source strength, there is no adjustable parameter by which the two parts of the spectrum can be shifted independently.

Tables I and II present the calculated pulse-height distributions to be expected for Ne^{19} and A^{35} positrons, normalized to a prescribed photopeak area in the manner just described. The integration was done for each of the four processes separately, and their individual contributions are included in the tables to show their relative importance. The values at a pulse height of 1.2 Mev are open to some doubt, because the corresponding response curve $f(k,V)$ was obtained by an uncertain extrapolation.

The comparison of theoretical and experimental distributions is shown in Figs. 6, 8, and 9. The theoret-

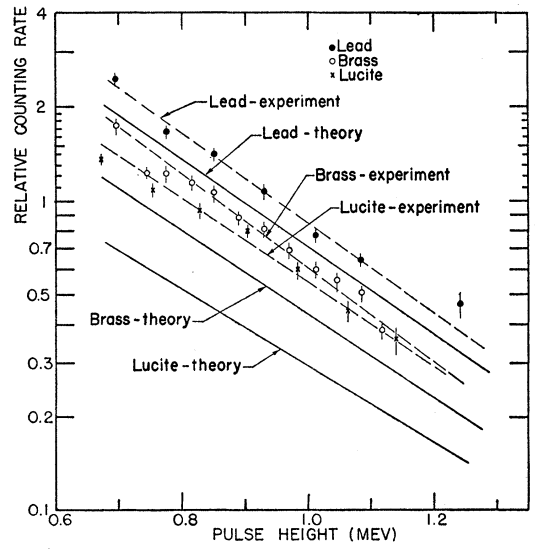

FIG. 9. Comparison of theoretical pulse-height distributions and data for 4.4-Mev A^{35} positrons annihilating in lead, brass, and Lucite.

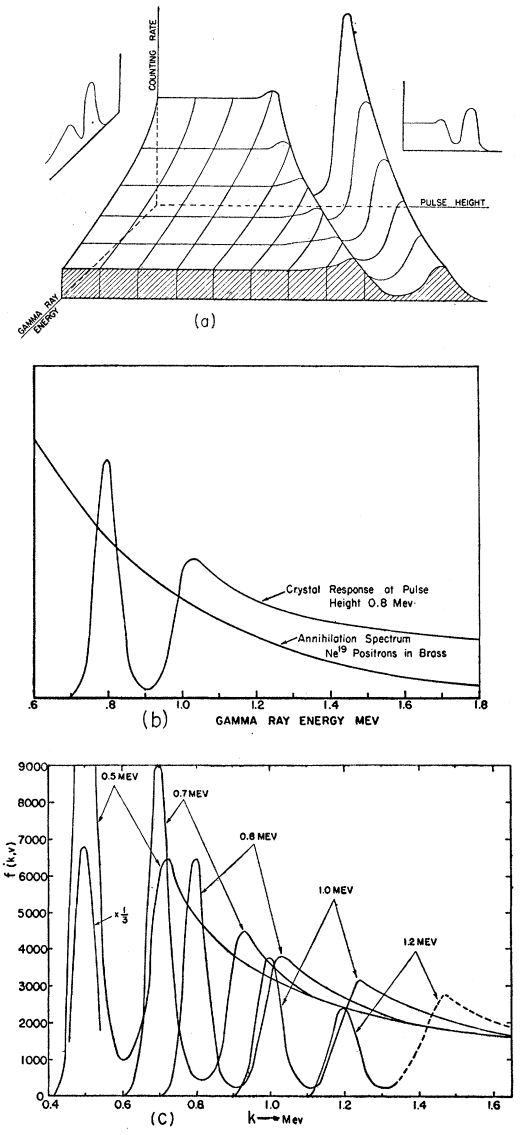

FIG. 10. (a) Three-dimensional plot of the NaI(Tl) crystal response $f(k,V)$ to monoenergetic γ rays as a function of γ-ray energy and pulse height. (b) A typical γ-ray spectrum and a cross section of $f(k,V)$ at a fixed pulse height, which is the weighting function for that pulse height. These two curves are multiplied and integrated over γ-ray energy k to give a single point on one of the pulse-height distributions. (c) Weighting function $f(k,V)$ as a function of k for various pulse heights V.

TABLE I. The calculated pulse-height distributions $\mathfrak{N}(V,\epsilon_0,Z)$ for Ne^{19} positrons. The $\mathfrak{N}$'s have been normalized to a prescribed 0.511-Mev photopeak area.

	V (Mev)	0.7	0.8	0.9	1.0	1.2
Lead	$\mathfrak{N}$	0.556	0.374	0.254	0.178	0.091
	$\mathfrak{N}_s$	0.033	0.033	0.034	0.033	0.030
	$\mathfrak{N}_{ob}$	0.124	0.062	0.032	0.016	0.004
	$\mathfrak{N}_{ib}$	0.007	0.004	0.003	0.002	0.001
	Total	0.72	0.47	0.32	0.23	0.126
Brass	$\mathfrak{N}$	0.465	0.315	0.217	0.151	0.079
	$\mathfrak{N}_s$	0.0004	0.0005	0.0005	0.0004	0.0004
	$\mathfrak{N}_{ob}$	0.038	0.019	0.010	0.005	0.001
	$\mathfrak{N}_{ib}$	0.007	0.004	0.003	0.002	0.001
	Total	0.51	0.34	0.23	0.158	0.081
Lucite	$\mathfrak{N}$	0.367	0.251	0.175	0.123	0.064
	$\mathfrak{N}_{ob}$	0.006	0.003	0.002	0.001	0.0002
	$\mathfrak{N}_{ib}$	0.007	0.004	0.003	0.002	0.001
	Total	0.38	0.26	0.180	0.126	0.065

ical curves shown in the figures are straight lines determined by the four calculated points in the interval from 0.7 to 1.0 Mev. The points calculated at 1.2 Mev lie above the lines, but the upward curvature implied by these points was not taken seriously in view of the uncertainty of the response function.

VI. APPROXIMATIONS AND EXPERIMENTAL ERRORS

The principal approximations made in the theoretical calculations and some uncertainties in the experimental data that have not yet been discussed are listed below.

1. Cross Sections

Both the annihilation and the bremsstrahlung cross sections are based on the Born approximation, whose accuracy in this energy region is very uncertain. In addition, the screened bremsstrahlung cross section has been approximated by a simple function of the initial and final positron energies.

2. Attenuation by Annihilation

The effect of attenuation by annihilation, mentioned in Sec. II, gives rise to two errors that partially compensate one another. Since a positron that annihilates in flight cannot make any further contributions to the γ-ray spectrum, the calculated spectra are too high by an amount varying from less than 8 percent for photons near 0.511 Mev to zero at the extremes of high- and low-frequency quanta. (The total probability of annihilation in flight is found to be 0.08 for A^{35} positrons in lead, and less than this for the other cases of present interest.) On the other hand, it is assumed in the normalization procedure that two quanta of 0.511 Mev are produced for each positron of the β spectrum, and the calculated photopeak area is therefore too high by an amount not exceeding 8 percent. After a normalization factor is applied to equalize the calculated and observed photopeak areas, the calculated hard spectrum will be too low by an amount increasing from zero near the photopeak to probably not more than 5 percent at a pulse height of 1 Mev.

3. Straggling

Further approximations are involved in taking account of the slowing-down of the positrons in the stopping material. Use of the average rate of energy loss in Eq. (3) to replace a path length dx by an energy interval $d\epsilon$ implies the disregard of energy straggling. In the presence of straggling, an expression for the average path length traversed in losing a fixed energy $d\epsilon$ ought to be used, if it were available, rather than the average energy lost in traversing a fixed path length dx.

4. Average Rate of Energy Loss

The mean ionization potential in Eq. (4) was taken to be 13.5 ev times Z. The Z for brass was taken as 29 (copper); for Lucite it was assumed that the stopping power is the sum of the stopping powers of the constituent elements. Since the chemical formula of Lucite is $|CH_2C(CH_3)COOCH_3|_N$, the effective Z is given by $54\ \ln Z = 30\ \ln 6 + 16\ \ln 8 + 8\ \ln 1$, or $Z = 5.0$.

In the calculations several small effects were neglected that influence the Z dependence of $d\epsilon/dx$. These are the density effect, the contribution of radiation loss to the stopping power, and the fact that I is not strictly proportional to Z. The calculations have not been redone with inclusion of these corrections, but their effect has been estimated by computing the corrected stopping power and comparing it with the uncorrected stopping power used in the calculations.

The values of I used in the corrected stopping power were taken from the proton range measurements of Bakker and Segrè[26] for metals and of Thompson[27] for

TABLE II. The calculated pulse-height distributions $\mathfrak{N}(V,\epsilon_0,Z)$ for A^{35} positrons. The $\mathfrak{N}$'s have been normalized to a prescribed 0.511-Mev photopeak area.

	V (Mev)	0.7	0.8	0.9	1.0	1.2
Lead	$\mathfrak{N}$	0.877	0.643	0.487	0.380	0.255
	$\mathfrak{N}_s$	0.054	0.055	0.055	0.054	0.053
	$\mathfrak{N}_{ob}$	0.959	0.614	0.409	0.280	0.145
	$\mathfrak{N}_{ib}$	0.029	0.020	0.015	0.011	0.007
	Total	1.92	1.33	0.97	0.73	0.46
Brass	$\mathfrak{N}$	0.764	0.559	0.426	0.335	0.224
	$\mathfrak{N}_s$	0.0007	0.0007	0.0007	0.0007	0.0007
	$\mathfrak{N}_{ob}$	0.296	0.192	0.127	0.088	0.045
	$\mathfrak{N}_{ib}$	0.029	0.020	0.015	0.011	0.007
	Total	1.09	0.77	0.57	0.44	0.28
Lucite	$\mathfrak{N}$	0.627	0.462	0.351	0.277	0.186
	$\mathfrak{N}_{ob}$	0.049	0.032	0.021	0.014	0.007
	$\mathfrak{N}_{ib}$	0.029	0.020	0.015	0.011	0.007
	Total	0.71	0.51	0.39	0.30	0.20

[26] C. J. Bakker and E. Segrè, Phys. Rev. **81**, 489 (1951).
[27] T. J. Thompson, University of California Radiation Laboratory Report No. 1910, 1952 (unpublished).

the constitutents of organic compounds. For lead, $I=758$ ev$=82\times9.25$ ev; for copper, $I=279$ ev$=29\times9.6$ ev; and for brass (two-thirds copper and one-third zinc) we have taken $I=285$ ev. These values are lower than any other measurements of I for lead and copper, but we have used them for consistency because Thompson's results are based on the Bakker-Segrè value for copper. According to Thompson's prescriptions, the value of I for Lucite is 59 ev$=5.0\times11.8$ ev, where account is taken of the fact that the polymerized form of methyl methacrylate has only one double bond per molecular unit.

The correction for density effect was taken from the formulas of Sternheimer.[28] His values for polystyrene should also be a good approximation for Lucite, because Lucite has 10 percent more electrons per unit volume but they are a little more tightly bound and give a weaker polarization per electron. An estimate of the contribution of radiation loss was obtained from the cross section (10) by an integration over photon energy. Radiation loss and density effect produce corrections of opposite sign, with density effect predominating in Lucite and radiation loss in lead and brass.

The new values of average energy loss differ from the values used in the calculations by factors that are fairly constant in brass and Lucite but change significantly over the higher energy range in lead. Averaged over the energy range of importance in the Ne^{19} calculations, the *reciprocal* of the corrected stopping power is equal to the reciprocal value used in the calculations multiplied by 0.99 in Lucite, 0.95 in brass, and 0.90 in lead. The corresponding ratios for A^{35} are 1.00, 0.94, and approximately 0.85. Since the reciprocal stopping power enters directly into every calculated photon spectrum, with the unimportant exception of inner bremsstrahlung, the product of a calculated pulse-height distribution and the appropriate one of the ratios just listed should give a good estimate of the pulse-height distribution that would be obtained from a calculation with the corrected stopping power. The resultant diminution of Z dependence would be in the right direction but not sufficiently marked to produce agreement between theory and experiment. The reduction in magnitude of the calculated counting rates in brass and lead would be in the wrong direction, except for the case of Ne^{19} positrons in lead.

The fact that the Bakker-Segrè ionization potentials are rather low does not alter these conclusions appreciably. If we take $I=29\times10.5$ ev as a probably better value for copper, and raise the other ionization potentials in the same proportion, all the ratios listed above are increased by 0.01.

5. End-Point Energies

The end-point energy of the A^{35} β spectrum was taken to be 4.40 Mev on the basis of two cloud-chamber determinations[29,30] that led to 4.38 ± 0.07 Mev and 4.41 ± 0.09 Mev. A larger value for the end-point energy would narrow the gap between the calculated and observed hard radiation spectra, but an increase sufficient to make a substantial improvement would produce a disagreement in the slopes. It would also violate the pattern of Coulomb energies in the mirror nuclei, since an end-point energy of 4.4 Mev corresponds to a Coulomb radius of $1.45A^{\frac{1}{3}}\times10^{-13}$ cm.

The end point of the Ne^{19} β spectrum was taken in the calculations to be 2.3 Mev on the basis of cloud chamber and particularly of absorption measurements.[25] However, a spectrometer measurement[31] has led to a value of 2.18 ± 0.03 Mev, and a threshold determination of the $F^{19}(p,n)Ne^{19}$ reaction[32] to an end-point value of 2.235 ± 0.005 Mev. In order to check the sensitivity of the calculated hard radiation spectrum to small changes in end-point energy and ionization potential, the calculations for Ne^{19} positrons in brass have been repeated with $I=10.5Z$ and $T_0=2.18$ Mev (instead of $13.5Z$ and 2.3 Mev). The new spectrum is lower than the original one by an amount increasing from 8 percent at a pulse height of 0.7 Mev to 15 percent at 1.2 Mev. The change in I accounts for difference of 4 to 3 percent, and the change in T_0 for the remainder. A spectrum calculated with $I=10.5Z$ and $T_0=2.235$ Mev would therefore be lower than the original one by an estimated 6 to 9 percent. The slope would be essentially the same and the agreement with experiment somewhat worse.

6. Absorption

A principal uncertainty in the data for which no correction has been attempted, as mentioned earlier, is the differential absorption of the γ rays in various parts of the annihilation spectrum by the walls of the brass gas chamber (lower-energy γ rays being more strongly absorbed than harder radiation). An experimental estimate of the magnitude of this effect indicated that the counting rates in the hard radiation NaI(Tl) spectrum should be reduced by less than 5 percent before being compared with the computed spectra. This correction is small because the geometry, from the standpoint of γ absorption, is "bad," so that the absorption of direct radiation is partially compensated by in-scattering.

7. Normalization

A second experimental uncertainty arises in the comparison of the calculations with the data. The method of normalizing the calculations to the data was, in essence, to equate the calculated fraction of radiation from positrons annihilating at rest multiplied by the

[28] R. M. Sternheimer, Phys. Rev. **88**, 851 (1952); **91**, 256 (1953).

[29] White, Creutz, Delsasso, and Wilson, Phys. Rev. **59**, 63 (1941).

[30] D. R. Elliott and L. P. D. King, Phys. Rev. **60**, 489 (1941).

[31] G. Schrank and J. R. Richardson, Phys. Rev. **86**, 248 (1952).

[32] Willard, Bair, Kington, Hahn, Snyder, and Green, Phys. Rev. **85**, 849 (1952). See also C. W. Li, Phys. Rev. **88**, 1038 (1952).

TABLE III. Estimated percentage corrections to calculated and experimental pulse-height distributions. The effects listed refer to those discussed under the same headings in Sec. VI. A positive sign indicates that the correction would raise the theoretical pulse-height distribution relative to the observed distribution.

	Ne^{19}			A^{35}		
	Lead	Brass	Lucite	Lead	Brass	Lucite
Attenuation by annihilation (at $V=0.9$ Mev)	+3%	+2.5%	+2%	+4%	+3.5%	+3%
Average energy loss	−9	−4	0	−14	−5	+1
End-point energy (at $V=0.9$ Mev)	−4	−4	−4	0	0	0
Absorption	+5	+5	+5	+5	+5	+5
Normalization	−4	−4	−4	−8	−8	−8
Total	−9%	−5%	−1%	−13%	−5%	+1%

spectrometer photopeak efficiency for 0.511-Mev γ rays to the corresponding quantity obtained from the data. The former was taken to be the area under the 0.511-Mev photopeak in the response function $2f(1,V)$, which, except for the attenuation effect discussed in subsection 2, is the desired quantity. Correspondingly, for the data, this quantity was taken to be the observed 0.511-Mev photopeak area. This area, however, contains an extraneous contribution from the photopeaks and Compton distributions of the hard radiation on which the 0.511-Mev photopeak is superimposed. Though it was not possible to obtain an accurate value for the necessary correction, it can be estimated that the result of including the correction would be to make the counting rates in the calculated hard radiation spectrum roughly 4 percent lower for Ne^{19} and 8 percent lower for A^{35}. Note that the *relative* heights of the spectra of Figs. 8 and 9 are independent of the normalization procedure.

8. Summary

The estimated corrections discussed in the preceding paragraphs are summarized in Table III, with the exception of the very uncertain errors in the cross sections and the effects of straggling. The estimates listed in the table are in some cases rather crude, but it is felt that they are correct in order of magnitude.

VII. DISCUSSION

The experimental points for annihilation of Ne^{19} positrons in brass are seen (Fig. 8) to be in satisfactory agreement with the calculated spectrum, although they tend to lie above it at the end nearer the photopeak. This tendency is emphasized by a comparison of the areas under the theoretical and experimental curves between 0.7 and 1.0 Mev. The theoretical area is approximately 0.85 as large as the experimental one (defined by a dashed line in Fig. 8), and the corrections estimated in Table III would lower this ratio to 0.80. On the other hand, the calculated curve for lead appears to be distinctly above the experimental points except near the photopeak, but the comparison of areas is more satisfactory in this case, the ratio being 1.05 without corrections and 0.95 with the corrections estimated in Table III.

The ratio of the counting rate in lead to that in brass is much less encouraging than consideration of either stopping material alone. The calculated ratio is approximately 1.4 and the experimental ratio only 1.15. This discrepancy in the dependence on the stopping material is even more marked in the comparison of Lucite and brass; they cannot be distinguished by looking at the experimental points, whereas the calculated curve for Lucite is lower than that for brass by a factor 0.8. Indeed, the calculated counting rates in Lucite are, on the average, only two-thirds as large as the observed rates. Bremsstrahlung and single-quantum annihilation are too small to be relevant to this discrepancy, which is well outside the experimental error. A possible lack of isotropy in the Lucite experiments might decrease the discrepancy slightly, but could hardly account for a major portion of it.

All three experimental pulse-height distributions for Ne^{19} positrons appear to decrease more rapidly with increasing pulse height than the theoretical ones. Lowering the end-point energy of the Fermi spectrum to 2.235 Mev in the calculations would reduce this difference in the slopes only a very little, and corrections for attenuation by annihilation and for possible energy-dependent absorption effects would both tend to increase it.

For A^{35} positrons, the slopes are in better agreement with experiment, but the theoretical curves fall always below the experimental ones. For lead, brass, and Lucite, the ratios of calculated to experimental counting rates are approximately 0.85, 0.65, and 0.5, respectively; the corrections from Table III would lower the first two ratios even further. The theoretical Z dependence is again conspicuously larger than the experimental, and the corrections of Table III, while now in the right direction, remove only a small part of the discrepancy.

It should be noted that the Z dependence of the calculated distributions can be traced directly back to the cross sections and to the rate of energy loss. It is not affected by the normalization procedure nor by the response curves of the γ-ray detector, because these are the same for every stopping material. The excessive Z dependence of the calculated distributions for A^{35} positrons comes chiefly from the contributions of bremsstrahlung, and therefore implies that Eq. (8) represents a serious overestimate of the bremsstrahlung cross section for positrons, perhaps by a factor as large as 2.

From the comparison of Ne^{19} positrons in Lucite and brass, where two-quantum annihilation is the only important source of γ rays, it appears that the $\ln I^2$ term of the stopping power produces a stronger Z

dependence than is observed. This conclusion still holds when the values of I are taken from proton data, instead of being assumed proportional to Z, and when corrections for the density effect are included. The same remarks apply with less force to the comparison of Ne^{19} positrons in lead and brass, since the logarithmic dependence of the stopping power would alone be sufficient to produce a lead-to-brass ratio of 1.17 (1.13 with the corrections of Table III), whereas the experimental ratio of 1.15 could be attributed entirely to the effects of bremsstrahlung and single-quantum annihilation.

If the conclusion is accepted that bremsstrahlung has been considerably overestimated in the calculations, it must also be recognized that a 50 percent reduction in the bremsstrahlung cross section would make the calculated pulse-height distributions for A^{35} positrons in lead and brass smaller than the experimental ones by factors of 0.5 to 0.7. This discrepancy would then be similar for A^{35} positrons in all three stopping materials, but would not be shared by the lower-energy positrons of Ne^{19} and C^{11}.

In summary, the experimental results support the γ-ray energy dependence of the two-quantum annihilation cross section and, in some cases, but not in others, the magnitude of the cross section. The agreement is satisfactory for Ne^{19} positrons stopping in brass or lead and for C^{11} positrons, but not for Ne^{19} positrons in Lucite. The disagreement for the higher-energy A^{35} positrons is large and has no apparent explanation, unless it is due to an inadequacy in the cross section in this energy region.

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A Precise Momentum Measurement of the F Line of Thorium B*†

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The momentum of the internal conversion electrons of the F line of thorium B has been compared with the momentum of electrons accelerated through a known potential difference. The comparison was made by means of an iron-free high-resolution solenoidal magnetic field beta-ray spectrometer. The value obtained for the F line is 1388.56±0.21 gauss-cm. Other useful calibration lines following the decay of thorium B have also been measured by comparison with the F line.

INTRODUCTION

UNTIL recently the most accurately known beta-ray momentum standard was Siegbahn's[1] measurement of the F internal conversion line in thorium B. The interest in precision beta- and gamma-ray spectroscopy received considerable stimulation by the accurate curved-crystal spectrometer wavelength determinations of nuclear gamma rays and annihilation radiation by DuMond and others.[2,3] The initial objectives of the present work[4] were to improve existing standards, and to endeavor to resolve an apparent inconsistency between the calculated and measured energy of annihilation radiation.[5] DuMond[6] pointed out that the discrepancy had been noted in earlier determinations[3] of the energy of annihilation radiation, and that it could be accounted for by an electron-positron mass difference of ~250 ev.

While the experiment discussed in this paper was in progress, several other careful energy measurements were reported. Lindström[7] determined the momentum of the F, I, and L internal conversion lines of thorium by a direct measurement of the deflecting magnetic field (in terms of the proton magnetic moment) and of the radius of curvature of the electron trajectories. A determination of the momentum of the A, F, and I lines by a different method was made by Craig.[8] In addition, other standard conversion lines and gamma rays have been measured with considerable accuracy.[9,10]

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