Positron Transmission and Scattering Measurements Using Superposition of Annihilation Line Shapes: Backscatter Coefficients*

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Backscatter coefficients have been measured for positron spectra emitted by radioactive sources and incident on 20 elements ranging from Li to U. The technique, based on recording the annihilation-γ-ray spectrum in a Ge (Li) detector, eliminates uncertainties encountered in methods where the positrons themselves are detected. The coefficient R for backscatter into 2π is found to vary with atomic number Z according to the relationship $R = 0.342 \ln Z - 0.146$.

I. INTRODUCTION

In this and in succeeding papers we report measurements on the transmission and backscattering of positrons emitted by radioactive sources and incident upon a range of elements extending over the entire Periodic Table. The primary motivation for this work arose from our need for basic data on positron penetration and reflection in studies of the trapping of positrons in metal defects such as dislocations and vacancies.

The technique employed here differs radically from the methods of previous workers in that the positrons themselves are not detected. Instead, we employ Ge (Li) spectroscopy of the annihilation γ rays, which were actually a nuisance factor in earlier works. The degree to which the energy distribution of these γ rays, centered at 511 keV, is Doppler broadened, reflects the electron momentum distribution in the material in which annihilation of the positron occurred. Thus, if positrons are brought to rest in more than one material, the fractions annihilating in the different materials may be deduced from the observed γ-ray spectrum. Provided that the detection efficiency is the same for annihilation γ rays arising in the various materials, this spectrum is simply the sum of components characteristic of each material. Further, it is even possible to restrict studies to a single material, since the annihilation-γ-ray spectra from different samples may be made characteristic by introducing a controlled number of defects into the lattice of each sample.

As a first stage in this work we have measured the backscatter or reflection coefficient $R(Z)$ for a range of elements extending from $Z = 3$ (lithium) to $Z = 92$ (uranium). In order to vary the positron energy, different radioactive sources were used. While a smooth relationship between $R(Z)$ and Z was found, the results did not support the square-root law suggested by the more limited data of earlier workers. The previous work, both experimental and theoretical, is reviewed in Sec. II and the present measurements on backscattering are described in Secs. III and IV. Conclusions are drawn and the next stage of investigation is indicated in Sec. V.

It should be made clear at the outset that our interest here is in a specific geometrical situation, which may at first sight appear rather restricted, but which is in fact the situation frequently prevailing in current efforts to employ positron annihilation in the study of metal defects. A list of references to such work can be found in Ref. 1. The geometry involves an essentially point radioactive source placed in contact with a sheet of the metal under study, the thickness of which is sufficient to ensure that any positron which is not reflected from the material is brought to rest within it. The backscatter coefficient $R(Z)$ is then defined simply as the fraction of those positrons incident on the surface that eventually annihilates outside the material. There is no dependence on angles of incidence or reflection since the definition effectively integrates over a solid angle of 2π for both. Further, the definition refers to the integrated spectrum of β energies emitted by the source and not to selected energies. Our review of previous work (Sec. II) is therefore concerned mainly with work involving integration over all energies emitted by radioactive sources. We do refer, however, to two geometries, viz., that described above and also the case where only those β's are counted which are backscattered into a cone of a half-angle considerably less than $\frac{1}{2}\pi$ and having its axis perpendicular to the face of the scatterer.

To illustrate the need for backscatter coefficients of the type defined here we cite current attempts by ourselves and others to measure vacancy formation energies E_f for various metals by studying the annihilation characteristics of positrons trapped in the vacancies. The positron method[1] appears to promise a much greater degree of precision and reliability than conventional techniques, provided that

solutions are found to certain problems. Whether the positron lifetime, the angular correlation, or the Doppler broadening of annihilation γ rays is employed as the observable, the experimental arrangement is the same. A sealed positron source is sandwiched between two identical metal samples and observations are made over a wide temperature range, typically from 20 to over 1000 °C. The finite thickness of the material necessary to encase the source safely at these temperatures results in serious systematic uncertainty; an unknown fraction of observed annihilation events occur in the source envelope material rather than in the material under study. Since reflection coefficients are typically 30–50%, a given positron may traverse the envelope several times, resulting in a non-negligible probability of absorption there. If the reflection coefficients for the envelope material and the metal under study are known, the requisite corrections can be made.

II. PREVIOUS WORK ON BACKSCATTER OF NUCLEAR β SPECTRA

In its passage through matter a positive or negative electron loses kinetic energy mainly through inelastic collisions with atomic electrons. Large deflections of the positron from its original path are due mainly to scattering by nuclei.

A single Coulomb scattering of an electron of given energy can of course be treated theoretically[2,3] without difficulty. The situation of interest here is far more complex. Nuclear β particles emerge from a radioactive source in all directions with a continuous distribution of energies up to some maximum W_0. Each particle can undergo a variety of interactions within any material on which it may impinge, and the β's eventually diffuse in all directions in the material. It is not surprising that there has been very little theoretical effort expended to calculate the fractions of positrons that eventually come to rest inside and outside a scatterer; most of the experimental work has been interpreted empirically.

We shall not review here the very early measurements on accelerated monoenergetic electrons and on β^- particles made before the advent of the Geiger–Mueller counter; the relevant references are given by Knop and Paul.[4]

The first extensive experimental effort is found in the work of Yaffe and Justus[5] and in two sequential investigations of Seliger.[6,7]

The former work and Seliger's first study both employed a very simple technique where an essentially weightless radioactive source deposited on very thin organic film (0.1 mg/cm^2) or aluminum leaf (0.22 mg/cm^2) was placed below the end window of a Geiger counter. Disks of various materials of sufficient thickness to ensure saturation backscattering were then placed directly below the source. In some experiments Seliger prepared aliquots of identical activity and size which were deposited directly onto the backscattering disks.

The percentage backscattering factor R measured by this method was found in both studies to be a smoothly increasing function of Z, levelling off at high Z, for both β^- and β^+ sources.

On first sight, however, the results for the dependence of $R(Z)$ on the maximum β energy W_0 appear to differ considerably. Yaffe and Justus, using the four β^- emitters listed in Table I, found a family of rather similar $R(Z)$ curves corresponding to different values of W_0; the backscattering increased markedly with W_0. Seliger[6] found no significant dependence on W_0 for his three β^- sources, also listed in Table I. He observed, however, that the backscattering factor for β^+ particles from a ^{22}Na source was consistently lower than the β^- value over the entire range of atomic number.

We shall not display these results graphically here. Since the two geometries doubtless differed, any intercomparison could be misleading.

The above evidence is insufficient to permit the conclusion that the two sets of results are in disagreement. The energy dependence observed by Yaffe and Justus appears to be most dramatic for the very-low-energy emitters ^{35}S and ^{60}Co; the difference between the ^{32}P and ^{106}Ru data is small. Seliger did not study any β^- emitters of comparably low energy, and so even if the effect is real it is unlikely that he would have observed it with the fairly high energies he used. However, Seliger's β^+ data, which are indeed low, were obtained with his lowest energy source.

One is therefore left with various possible explanations of these early results. The observed effects may be attributable entirely to energy dependence. There may be no energy dependence but a difference in backscattering, dependent upon charge. This would imply the existence of a systematic error in the Yaffe and Justus work, but the present authors can certainly discern no errors in this careful work. Finally, of course, both effects may contribute.

TABLE I. β emitters studied in Refs. 5–7.

Yaffe and Justus (Ref. 5)		Seliger (Refs. 6 and 7)	
Emitter	W_0 (keV)	Emitter	W_0 (keV)
^{35}S (β^-)	167	^{131}I (β^-)	608 (87%)
			330 (9%)
^{60}Co (β^-)	314	^{210}Bi (β^-)	1160
^{32}P (β^-)	1710	^{32}P (β^-)	1710
^{106}Ru/^{106}Rh	2800 (20%)	^{22}Na (β^+)	545
(β^-)	3900 (80%)		

Seliger, apparently unaware of the β^- results of Yaffe and Justus, suggested that the reason for his β^+-β^- difference might lie in the fact that a relativistic treatment of Coulomb scattering shows an increase for β^- scattering and a decrease for β^+ scattering, as compared to the Rutherford formula. To investigate the β^--β^+ difference further, he devised a rather elegant experimental technique[7] which permitted him to estimate the effects of positron annihilation, a source of systematic error not amenable to measurement in his first experiment on ^{22}Na.

The absolute strength N_0 of ^{22}Na and ^{32}P sources deposited on thin polystyrene films was measured by placing the films in turn at the center of a 4π proportional counter. The outer spherical electrode was divided into two equal parts by an insulator. Measurement of the backscatter coefficient R into 2π sr was then accomplished by placing a reflecting foil under the source and measuring the counting rate in the upper hemisphere. R was deduced from the enhancement of this rate over N_0.

In the case of ^{22}Na, a spurious count in the upper hemisphere could result from detection of an annihilation γ ray from a positron which had come to rest in the reflector. The magnitude of this effect was estimated from the counting rate in the lower hemisphere. A second correction necessary only for ^{22}Na, whose positrons are of relatively low energy, was that for positron absorption in the plastic foils enclosing the source.

Seliger's results again exhibited a marked difference in the backscatter coefficients measured for positive and negative electrons, the values for β^- exceeding those for β^+ by some 30% at high Z. Miller[8] attempted to render Seliger's proposed explanation of this observation somewhat more quantitative. The difference in relativistic elastic scattering cross sections for positive and negative electrons amounts to a factor of 4 for 90° scattering at 1.7 MeV; it is not obvious, however, that it is totally responsible for the observed effect, since at lower energies and small angles the cross sections tend towards equality. Multiple small-angle scatterings are in fact a major cause of backscattering from materials of high atomic number.

Miller made an estimate of the ratio of backscatter coefficients at $Z = 80$ using a version of a neutron transport calculation of Bothe.[9] Miller used theoretical relativistic electron scattering cross sections to estimate an electron "scattering length" and also adopted a value of $R = 0.030$ cm for the electron range in mercury. The result of this calculation was 1.16, in fair agreement with Seliger's observation. Miller warned, however, that the calculation was of a sufficiently *ad hoc* nature that no definite conclusion should be drawn from it.

Further experimental work was carried out by various authors, all of whom used straightforward experimental arrangements, where a Geiger counter recorded the intensity of electrons and positrons backscattered from various materials into defined solid angles generally considerably less than 2π. Muller[10] studied 32 elements in the range $4 < Z < 83$, using the 2 MeV β^- spectrum from ^{90}Y. His results indicated that the relative backscattering factor R was a discontinuous function of Z, although strictly linear in Z within each portion of the periodic system. When the linear equations relating R to Z within each period were solved simultaneously, discontinuities become apparent at $Z = 10$, 18, 36, and 54, corresponding to the rare gases which terminate the periods. In contradiction to these results, both Danguy and Quivy,[11] using β^- particles and Cambieri and Pappalordo,[12] using β^+ particles, observed a smooth dependence proportional to the square root of Z in the range $13 < Z < 82$.

Experiments of this type on positrons are subject to error due to the detection of γ rays from positron annihilation in the scatterer or in surrounding materials. Moreover, they sample only a fraction of the total backscattering into 2π, which is the quantity of interest here.

Seliger's second study overcame these problems in an elegant way, but his work was restricted to only five values of Z. More recently, novel solutions have been introduced by Bisi and Braicovich[13] and independently by Finley, McKee, and MacKenzie.[14] Both of these groups realized that one solution to the first problem was to ensure that all positrons were annihilated and to employ the characteristics of the annihilation γ radiation to identify the annihilation site as being either inside or outside the backscatterer. To ensure annihilation of all positrons, the radioactive source had to be sandwiched between two foils of sufficient thickness to ensure total absorption. This automatically solved the second problem since all positrons backscattered from the material constituting one side of the sandwich were then incident on the different material of the other side.

The quantity measured was the lifetime of the positrons. One side of the sandwich was the metal of interest, in which due to the high electron density the lifetime is short (of the order of 0.15–0.5 nsec, depending on the metal). The other side of the sandwich was a sheet of Teflon, a material which, in common with other organic solids, displays several lifetime components arising from various processes which need not be discussed in detail here. The salient point is that a long-lifetime component of some 2–4 nsec, due to pickoff annihilation of positrons bound in orthopositronium, is

characteristic only of the Teflon and can thus serve as a measure of the fraction of positrons annihilating there. The backscatter coefficient R of the metal may then be deduced from that fraction.

With this method, Bisi and Braicovich observed the dependence

$$R = (0.0593 \pm 0.0019) Z^{1/2}. \quad (1)$$

However, their results were analyzed on the basis of there being two lifetimes in Teflon, a "short" lifetime of about 0.3 nsec and the long lifetime discussed above. Several workers have since demonstrated the existence of an intermediate component ($\simeq 1$ nsec) responsible for about 30% of all annihilations in Teflon. This observation casts some doubts on the validity of Eq. (1).

In their independent measurements using the lifetime method, Finley, McKee, and MacKenzie did take account of the intermediate component. They found the dependence

$$R = (0.054 \pm 0.001) Z^{1/2}, \quad (2)$$

up to $Z = 50$, but at higher Z, the reflection coefficients fell below the $Z^{1/2}$ relationship. Insufficient points were available to encourage attempts towards a functional fit of wider validity.

It is clear from the foregoing that very different results emerge from the various techniques. It appears, however, that the index n in the $R = aZ^n$ relationship is likely to be of the order of 0.5. While the lifetime measurements might initially be regarded as the more reliable, they are subject to several disadvantages, apart from the obvious ones of complexity of fast timing electronics and lengthy duration of coincidence measurements. Analysis of complex lifetime spectra is not straightforward and there is still not agreement as to the number of lifetime components for a polymer.[15]

There is no physical reason to expect an exact square-root relationship between R and Z. It is fortunate that a degree of simplicity is introduced by the energy independence of R. Blanchard and Fano[16] have shown that beams of electrons traversing a material lose their original sense of direction at the same fractional loss of energy. Thus for a given Z, the same fraction of each should undergo reflection, and the greater penetration of the more energetic particles before losing their sense of direction is compensated by the greater energy they retain to retraverse the material to the surface.

Seliger suggested that the Z dependence of R is due to the fact that the ratio of elastic to inelastic scattering is proportional to Z. In his view, at high Z an electron or positron would, therefore, lose its sense of direction due to nuclear scattering encounters with much less energy loss (the loss being due to inelastic collisions with electrons) and hence much nearer to the surface than at low Z. There would therefore be a greater probability of escape from a high-Z material.

Although it has been emphasized earlier that work on backscattering of monoenergetic electrons at specified angles of incidence and reflection refers to a very different and less complex situation than that of interest here, some recent studies in this area should be cited. For example, Frank[17] employed monoenergetic electron beams from a betatron to study the distribution in angle and energy of electrons backscattered from various materials. Frank deduced the backscatter coefficient into 2π for a 1.75-MeV beam incident on a surface at 90° by integrating his measured angular distribution curves; his result will be compared later with that determined by the present workers for a ^{68}Ge positron source giving a diffuse incident angular distribution and a continuum of energies up to 1.8 MeV.

Archard[18] attempted to place the theory for negative electrons incident perpendicular to a target on a quantitative basis. He suggested that at low Z the observed backscattering is predominantly due to single-elastic-scattering collisions. As Z rises, the penetration into the material decreases and more electrons are able to diffuse out of the surface, having lost their original sense of direction after a large number of collisions. At high Z, electrons are diffused almost immediately and there is little chance of single elastic scattering. Archard calculated the $R(Z)$ for these two mechanisms in the extremes of low and high Z and employed a weighted average at intermediate Z. His predictions are discussed further in Sec. V.

More recently, Tillmann[19] has performed Monte Carlo calculations of backscattered energy spectra and angular distributions for monoenergetic electrons of 0.5, 1.0, and 2.0 MeV incident at various angles on various materials. Backscatter coefficients are found to fit a $Z^{1/2}$ dependence for $Z > 13$. The coefficients are strongly dependent on the angle of incidence, and a decrease in backscattering is predicted as the energy is increased from 1 to 2 MeV. Rigorous comparison of this theory with our data would require that the Monte Carlo treatment be extended such as to effectively integrate over incident angles and energies.

III. BASIS OF PRESENT MEASUREMENTS

It is widely accepted that positrons, upon entering a metal, are rapidly thermalized ($\simeq 10^{-12}$ sec) before undergoing annihilation with conduction or core electrons. The momentum of the electron-positron pair immediately prior to annihilation is thus determined almost entirely by the electron-momentum distribution of the material. A γ-ray spectrometer viewing the metal will then record a γ-energy distribution centered at $0.5(m_{e^-} + m_{e^+})$

= 511.002 keV and having a Doppler broadening which reflects the electron-momentum distribution. Currently available Ge (Li) detectors respond to a monoenergetic photon line at this energy with an approximately Gaussian pulse-height distribution whose resolution [full width at half-maximum (FWHM)] is about 1.5 keV. This figure is some two to three times smaller than the spread of the annihilation line, as is shown in Fig. 1, where the instrumental resolution function has been determined by recording the peak from the 514-keV nuclear γ ray emitted by ^{85}Sr, and the annihilation line is that of metallic copper.

In spite of the fact that it is only very recently that the energy resolution exhibited in Fig. 1 has been attained, it is not likely that Ge (Li) spectroscopy will become a widely used tool for the measurement of electron-momentum distributions in solids. The long-established technique of measuring the angular correlation between two annihilation γ rays accomplishes this task with almost an order of magnitude better resolution. However, the much greater rate of data accumulation possible with the Ge (Li) technique opens up the possibility of studies of phenomena that result in observable changes in the line shape. It has been demon-

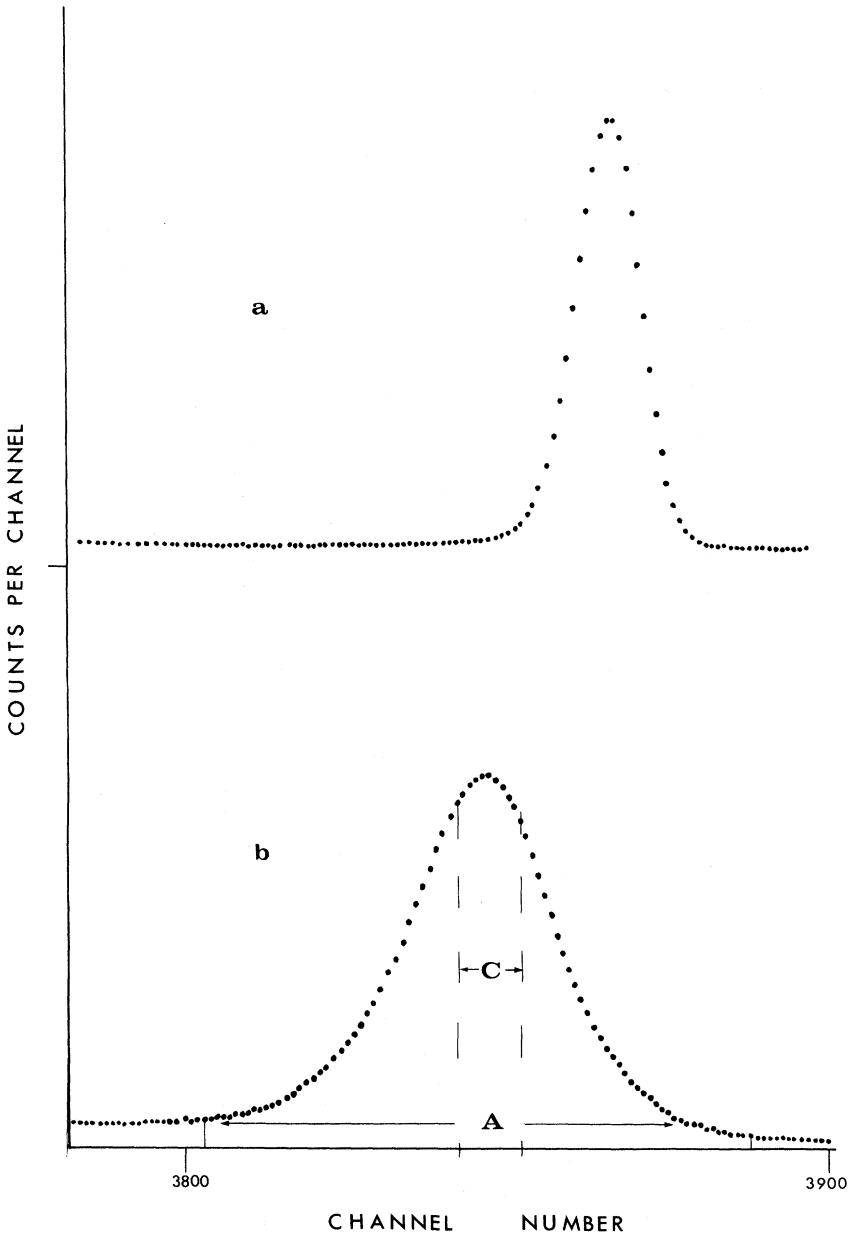

FIG. 1. Pulse-height spectra due to γ radiation recorded with Ge (Li) detector: (a) 514-keV γ-ray peak from ^{85}Sr decay; (b) 511-keV peak due to positron annihilation in copper. The ratio of areas C/A is defined as the "line-shape parameter" Q.

strated[20] that the presence of dislocations and vacancies results in positron trapping. Since the trapped and untrapped positrons sample different electron-momentum distributions, the Ge (Li) line shape varies with the defect density. Line-shape measurements have already been applied to a variety of fundamental and applied metallurgical problems,[21] and it is in fact this work that has pointed up the need for the basic data on ranges and backscattering coefficients under discussion here.

In order to characterize the line shape observed when a positron source is enclosed between two sheets of a given metal, we define a "line-shape parameter" Q as the ratio of the number of pulses C falling in a defined group of channels in the central region of the peak to the total number of pulses A. Since the respective probabilities of any one γ ray giving rise to a pulse in the inner and outer regions are C/A and $1-C/A$, the quantity Q obeys the statistics of a binomial distribution. The channel groups used here to define Q are shown in Fig. 1; these groups were defined in such a way as to ensure the maximum possible variation of Q values among the metals of interest, while ensuring reproducibility for any one metal.

If the source is sandwiched between two different metals the value of Q observed will be a linear combination of the two independent Q values, weighted according to the fraction of the total number of positrons that annihilate in each metal.

In our experiment, Q is measured for three successive sandwich configurations as shown in Fig. 2. The first sandwich consists of two foils of the metal of interest X; the second consists of two foils of a standard metal S of supposedly a known backscatter coefficient; and the third consists of a composite of the two metals M. For the last case, P is defined as the fraction of positrons annihilating in X. Obviously then

$$Q_M = PQ_X + (1-P)Q_S,$$

so that

$$P = Q_S - Q_M / Q_S - Q_X. \tag{3}$$

If P is to be measured precisely, the standard metal chosen should have either as large or as small a Q value as possible.

It is a simple matter to show, by summing an infinite series of reflections under the assumption that the reflection coefficients R_X and R_S are energy independent, that

$$P = (1+R_X)(1+R_S)/2(1-R_XR_S) \tag{4}$$

so that

$$R_X = (1+R_S-2P)/(1+R_S-2PR_S). \tag{5}$$

The desired reflection coefficient is then obtained from the three measurements of Q, providing a value for the standard is assumed.

The error due to counting statistics may also be derived in a straightforward fashion. For a binomial distribution of the quantity $Q = C/A$, the standard deviations in the measured quantities A and C are, respectively, $A^{1/2}$ and $C^{1/2}(1-C/A)^{1/2}$.

Some points of uncertainty must also be considered here. It has been shown[1,21] that if low-energy positrons, such as those emitted by ^{22}Na ($W_0 = 0.54$ MeV), are incident on metallic surfaces prepared by different methods, then the line shape varies dramatically, the line-shape parameter changing monotonically with the degree of damage. All surfaces must therefore be carefully polished and etched before using. Errors from surface effects can be minimized by employing more penetrating positrons, such as those emitted in the decay ^{68}Ge $\rightarrow$ ^{68}Ga $\rightarrow$ ^{68}Zn ($W_0 = 1.8$ MeV).

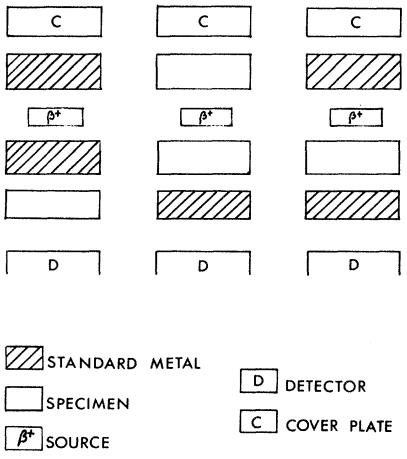

FIG. 2. Positron source configurations for annihilation-γ-ray line-shape measurements.

IV. EXPERIMENTAL PROCEDURE

Radioactive sources of a few microcuries activity of ^{68}Ge and ^{22}Na were prepared by droplet evaporation on 0.8 mil Kapton foil; the sources were covered by the same thickness of Kapton.

The Ge (Li) spectrometer used to record the annihilation line shapes was a Nuclear Diodes coaxial device (volume 20 cm^3) with a cooled FET preamplifier. It was employed in conjunction with a model No. TC202BLR amplifier and a Nuclear Data model No. 2200 pulse-height analyzer operated with conversion gain 4096. This system exhibited an energy resolution of 1.50 keV (FWHM) at 514 keV.

It is extremely important that small drifts in the electronic system be minimized since these would wipe out completely the relatively small changes in line shape of interest here. Accordingly, a dig-

ital stabilizer was used. Gain stabilization was accomplished by monitoring the 511-keV peak and zero stabilization by monitoring the 122-keV γ ray from a weak ^{57}Co source attached permanently to the detector.

Samples of a wide variety of metals ranging from lithium ($Z = 3$) to uranium ($Z = 92$) were prepared in the form of disks of diameter of about 10 mm; the thickness varied from one metal to another but was sufficient to ensure complete absorption of ^{68}Ge positrons. Magnesium was chosen as a standard (X) since its line shape was the narrowest observed among those metals which could be handled conveniently. All samples except lithium were annealed just below the melting point, allowed to cool slowly, polished with successively finer grades of emery cloth, lapped and etched before commencement of measurements.

The quantity P was measured for a number of metals with Mg as standard; the results are given in Table II(A). In cases such as lead where rapid surface oxidation occurred, the sample was placed in vacuum immediately after preparation and the γ rays emerged from the vacuum chamber through a 10-mil beryllium window. Source-detector separation was adjusted to obtain 5000 (± 2%) counts per sec in each case. The effect of counting-rate variations of 20% was investigated at the outset and it was found that no observable changes in the line-shape parameter resulted. Since for the three sim-

TABLE II. Experimental data for annihilation fraction P and derived backscatter coefficient R.

	Standard	Sample	Z	Positron source	P	R
(A)	Mg	C	6	Ge	0.5534 ± 0.0069	0.137 ± 0.014
		C		Ge	0.5533 ± 0.0069	0.137 ± 0.014
		Fe	26	Ge	0.4405 ± 0.0037	0.349 ± 0.007
		Fe		Ge	0.4509 ± 0.0038	0.330 ± 0.007
		Ni	28	Ge	0.4362 ± 0.0036	0.356 ± 0.007
		Cu	29	Ge	0.4375 ± 0.0039	0.354 ± 0.008
		Cu		Na	0.4210 ± 0.0051	0.384 ± 0.010
		Cu		Na	0.4462 ± 0.0023	0.339 ± 0.005
		Cu (rolled)		Ge	0.4244 ± 0.0049	0.377 ± 0.009
		Cu		Ge	0.4453 ± 0.0024	0.340 ± 0.005
		Cu		Ge	0.4461 ± 0.0029	0.339 ± 0.006
		Zn	30	Ge	0.4397 ± 0.0061	0.350 ± 0.012
		Ge	32	Ge	0.4280 ± 0.0105	0.371 ± 0.020
		Mo	42	Ge	0.4135 ± 0.0040	0.397 ± 0.008
		Mo		Ge	0.3947 ± 0.0028	0.429 ± 0.005
		Mo		Na	0.4065 ± 0.0057	0.409 ± 0.011
		Ag	47	Ge	0.4054 ± 0.0043	0.411 ± 0.008
		Ag		Ge	0.3912 ± 0.0042	0.435 ± 0.008
		Cd		Ge	0.3889 ± 0.0065	0.439 ± 0.012
		In	49	Ge	0.3986 ± 0.0093	0.422 ± 0.018
		Sn	50	Ge	0.3683 ± 0.0097	0.474 ± 0.018
		W	74	Ge	0.3446 ± 0.0042	0.513 ± 0.008
		W		Ge	0.3556 ± 0.0041	0.494 ± 0.008
		W		Na	0.3643 ± 0.0058	0.480 ± 0.011
		Au	79	Ge	0.3496 ± 0.0037	0.504 ± 0.007
		Pb	82	Ge	0.3423 ± 0.0064	0.516 ± 0.012
		Bi	83	Ge	0.3157 ± 0.0119	0.559 ± 0.022
		Bi		Ge	0.3542 ± 0.0084	0.497 ± 0.016
		U	92	Ge	0.3413 ± 0.0036	0.518 ± 0.007
(B)	Cu	Al	13	Ge	0.5695 ± 0.0062	0.228 ± 0.024
		Al (rolled)		Ge	0.5749 ± 0.0057	0.218 ± 0.024
		Ge	32	Ge	0.4982 ± 0.0064	0.359 ± 0.024
		In	49	Ge	0.4485 ± 0.0063	0.442 ± 0.024
		Sn	50	Ge	0.4470 ± 0.0063	0.445 ± 0.024
		Ho	67	Na	0.4309 ± 0.0056	0.471 ± 0.023
		Yb	70	Na	0.4297 ± 0.0045	0.473 ± 0.022
		Pb	82	Ge	0.3975 ± 0.0076	0.523 ± 0.025
		Bi	83	Ge	0.4149 ± 0.0053	0.496 ± 0.023
(C)	Mo	Li	3	Ge	0.6923 ± 0.0035	0.032 ± 0.018

ple sandwich configurations, the 511-keV γ rays would traverse different materials and hence undergo slightly different scattering en route to the Ge (Li) detector, compensating metal disks were employed to eliminate this variable factor as shown in Fig. 2. The duration of a single line-shape-parameter determination was generally either 1 or 2 h.

It was observed that for certain metals, e.g., the rare earths, the line-shape parameter Q differed only slightly from the Q value of Mg; as a result the P value had a large uncertainty. Since the line-shape of Cu was extremely wide, this metal was adopted as a secondary standard. Its reflection coefficient was obtained very accurately in terms of the primary Mg coefficient by performing six Cu/Mg runs of longer duration than the 1 or 2 h normally employed.

The metals listed in Table II (B) were then run with copper as standard. The rare-earth foils available to us were, however, insufficiently thick to absorb completely the positrons of ^{68}Ge, and in these cases a ^{22}Na source was used. To ascertain whether or not serious surface problems might arise with ^{22}Na, several of the comparisons with Mg were repeated; the P values were in good agreement with those obtained using ^{68}Ge. Several of the Cu comparisons were then performed using ^{22}Na.

In one particular case, that of lithium, molybdenum was employed as standard, its reflection coefficient having been measured precisely in terms of that of Mg.

The various runs were not performed in any specific order and measurements on various metals were repeated at random. Only once was a discrepancy outside the range of statistical error suspected; in the case of bismuth one of the three P values was much lower than the others and departed from the trend in this region. No anomalous behavior is suspected, the bismuth specimen being rather unsatisfactory insofar as it was prepared by compressing bismuth chips.

Since a preliminary extension of this work to measurements of positron transmission in layered media had yielded some apparently anomalous results [22] for rolled foils containing dislocations, it was decided to conclude the present studies by measuring the backscatter coefficient for such foils. Accordingly, samples of copper and aluminum were rolled heavily so that all positrons would be trapped in dislocations before annihilating; standards of annealed magnesium and copper were employed, respectively. The results for these defective samples were in good agreement with those for annealed samples.

V. DISCUSSION OF RESULTS

In order to convert the data given in Table II into backscatter coefficients, a value must be adopted for the backscatter coefficient of the primary standard, magnesium. This degree of arbitrariness appears to constitute the only major disadvantage of the technique.

It would seem most sensible to adopt a value obtained using 2π geometry. This limits the choice to the value obtained in the 4π counter work of Seliger[7] or to that obtained in the lifetime work of Finley.[14] Since Archard's theoretical work[18] is for negative electrons and the different reflection of β^- and β^+ is not refuted, the possibility of normalizing our data to that of Archard at one value of Z and then assessing the goodness of fit over the Periodic Table does not arise.

Seliger's plot of R vs Z is drawn through six points, measured for lucite, aluminum, copper, silver, platinum, and lead. The value read from it for Mg lies between 0.27 and 0.28. Finley, McKee, and MacKenzie studied only four elements—aluminum, copper, indium, and lead—and their equation of best fit yields a value of 0.19 for Mg. This disagreement emphasizes the need for a reliable absolute measurement of backscatter coefficient to which our results can be normalized.

From the experimentalist's point of view, however, this is not a serious situation. The quantity generally required in experimental work is not the backscatter coefficient, but the quantity P which yields the fractions of positrons annihilating in the two different materials placed in contact. Since this is the primary quantity measured here, we list the experimental values of P in Table II. In the several cases where more than one measurement was performed on a given metal, we have chosen to list the results of all measurements. The purpose of this is to permit assessment of the degree of reproducibility afforded by the Ge (Li) technique. The errors will be discussed below.

With a value of $R = 0.24$ for the primary magnesium standard, a first set of backscatter coefficients was derived for the metals listed in Table II(A); for these the purely statistical errors range from about 1 to 5%. A mean value of R was calculated from the copper data and employed in analysis of the data of Table II (B), obtained using copper as standard. Similarly, a mean value was calculated for molybdenum in order to analyze the measurement on lithium, which used molybdenum as standard. The backscatter coefficients listed in Table II(B) have statistical errors ranging from 4.6 to 6.8%, the increase being due to the propagation of the uncertainty in the secondary copper standard.

Although the derived quantity R is of less direct value to the experimenter, it is the quantity that would be calculated from any scattering theory. Attempts

were therefore made to find the functional relationship between the R values and Z.

Various previous workers using both a 2π geometry identical to ours and other arrangements which detected only those positrons scattered in a cone of half-angle considerably less than $\frac{1}{2}\pi$ observed a $Z^{1/2}$ dependence. A least-squares linear fit was therefore made to the data of Table II using the logarithms of the quantities R and Z as ordinate and abscissa. The best fit was given by the equation

$$R = (0.101 \pm 0.003) Z^{(0.370 \pm 0.008)}, \quad (6)$$

which is represented by the continuous curve in Fig. 3. The dashed curve in the figure is that calculated by Archard[18] for electrons and is included only for general interest.

In this first comparison of our data with the previously used empirical formula $R = aZ^n$ and the simple theory, we have chosen to show all data points in the figure in preference to employing an average for each metal. The scatter can be attributed to statistical errors, which have been discussed previously and are given in Table II, to instrumental drifts, and to sample surface effects.

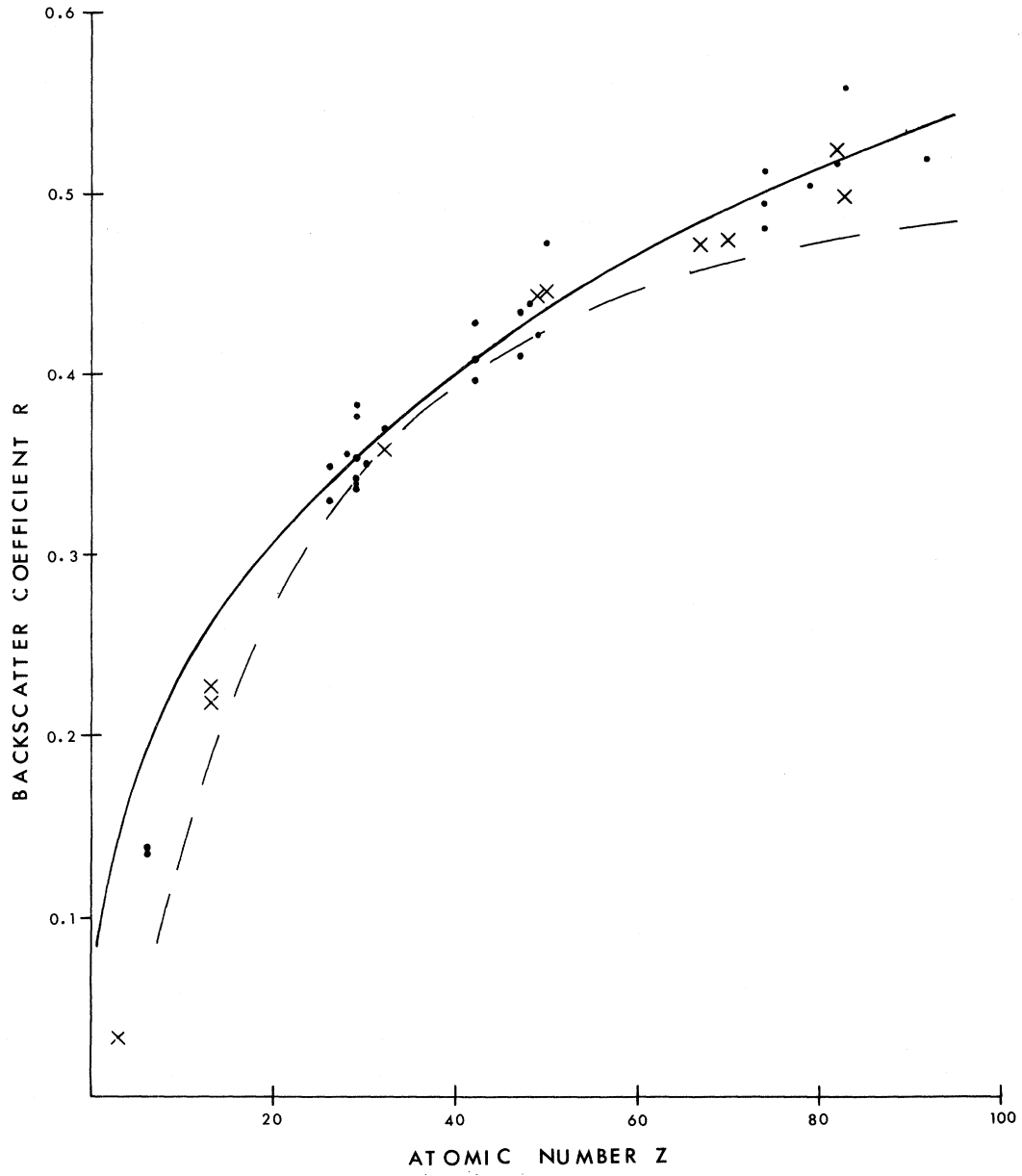

FIG. 3. Experimental data and best fit (curve) to $R = aZ^n$ for positron backscatter coefficient R as a function of atomic number Z. The dashed curve represents the values calculated for electrons by Archard's theory.

Although the pulse-height analysis system is stabilized, small residual gain shifts would cause changes in the line-shape parameter, especially for narrow peaks. Similarly, surface effects such as progressive oxidation would affect the line-shape parameter, generally causing a decrease in its value. The data indicate, however, that the latter two effects are small, counting statistics being sufficient to explain most of the observed scatter.

The main conclusion to be drawn is that our data do not confirm the $Z^{1/2}$ relationship suggested by earlier workers. This is not surprising since, as indicated earlier, there is no physical reason for the existence of such a simple relationship. In fact, Finley, McKee, and MacKenzie had already pointed out that while a $Z^{1/2}$ curve fitted their data at low and medium Z, there was evidence that the data at high Z fell below the values predicted by such a relationship.

While the fit afforded by Eq. (6) is fairly good in the region $Z > 25$, it clearly overestimates R for lower values of Z. Since inner-shell electrons are less important in causing scattering than are outer electrons, it might be argued that replacement of Z in Eq. (6) by $Z - \sigma$, where σ takes a value of about 3 or 4, would be appropriate. If this is done the fit remains good above $Z = 25$ and is improved at low-Z values. However, the zero value of R that results at $Z = \sigma$ indicates that this approach is physically incorrect.

Two further attempts to obtain a functional relationship are shown in Fig. 4, where for simplicity the weighted-mean-experimental values of R are now used. The function

$$R = 0.52\,(1 - e^{-0.045Z}), \tag{7}$$

chosen to yield a saturation effect at high Z, gives a fairly good fit at both low and high Z but badly overestimates R in the region $30 < Z < 70$. A much better fit over the entire region is afforded by the relationship

$$R = 0.342 \log_{10} Z - 0.146\,, \tag{8}$$

where the parameters have been obtained by a least-squares linear fit. This does not result in such a rapid saturation at high atomic number.

Although good fits could also be obtained with polynomials of sixth and seventh order, Eq. (8) is

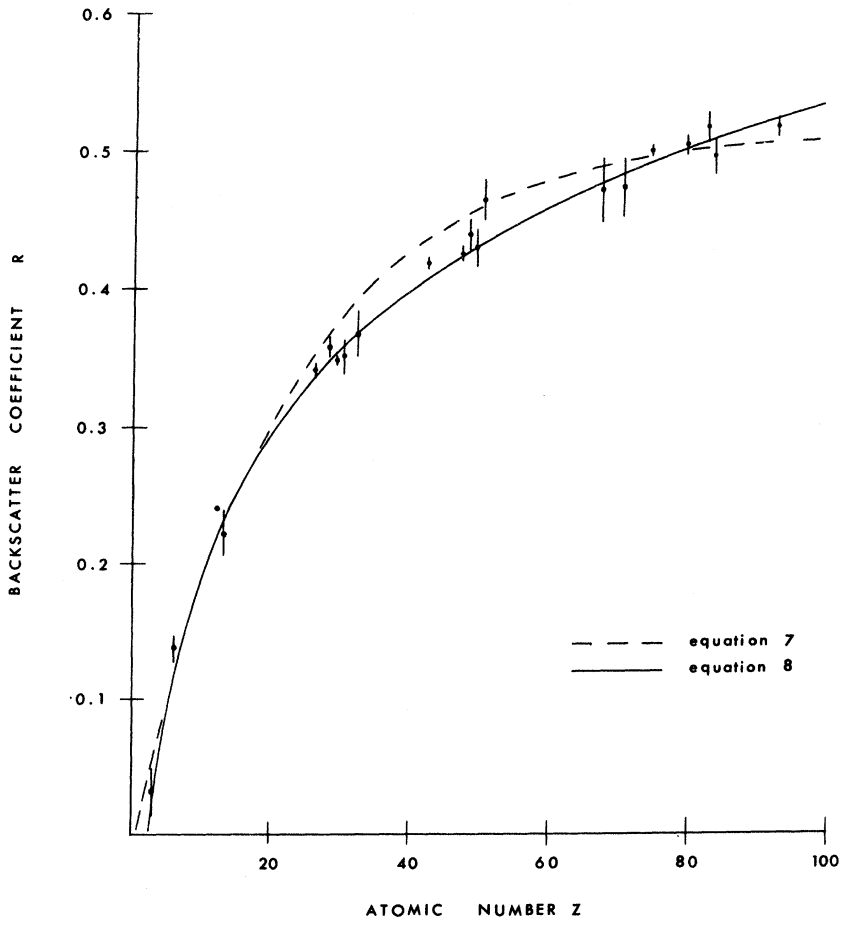

FIG. 4. Comparison of exponential (7) and logarithmic (8) fits to the weighted-mean-experimental data points. The best fit is provided by the full curve representing $R = 0.342 \log_{10} Z - 0.146$.

satisfactory for all practical purposes. It permits calculation of the backscatter coefficient for any element of interest with an uncertainty which is generally less than 5%, except at very low atomic number where experimental data are rather limited.

It must be recalled however, that the value of R adopted for Mg is not necessarily exact; in the absence of a precise absolute value for Mg, a systematic error amounting to about 6% at medium Z must be added to this uncertainty. Attempts to devise a method for an absolute measurement on Mg are now desirable, since if R (Mg) were known precisely, the present data would allow a stringent comparison with theory.

It is interesting to note that at medium Z our positron data agree rather well with the simple theory of Archard[18] for normally incident monoenergetic electrons; the theoretical curve has, however, a more pronounced curvature and so underestimates the data at low and high Z.

Since our data are for positrons only, we can neither uphold nor refute Seliger's original observation of smaller backscattering factors for positrons than for electrons. If Archard's theory were strictly applicable though, our data would not uphold Seliger's conclusion.

As mentioned earlier, no difference was observed in the results obtained with ^{22}Na and ^{68}Ge, whose maximum energies are very different. This is a rather interesting result in that it contradicts the observations of Yaffe and Justus[5] on β^- particles of differing energy. However, a definite conclusion should not be drawn since we have not yet employed a β^+ source of energy similar to the very low energies obtained from Yaffe's ^{35}S and ^{60}Co sources. Tillmann's Monte Carlo calculations[19] indicated that backscattering should decrease with increasing energy above 1 MeV for the special case of a monoenergetic normally incident beam. Clearly, therefore, the most useful comparison would be with an extension of Tillman's calculation, the latter involving integration over all incident angles and energies.

Finally, it may be claimed that the main objective of this work, measurement of the 2π backscatter coefficient over a wide range of Z for the two radioactive sources most widely used in positron-trapping studies of metal defects, has been achieved. The amount and quality of the data attests to the rapidity and reproducibility of the technique of superposition of annihilation line shapes. In the next stage of this program, the technique will be employed to study the depth distribution of positrons annihilating in various solids.

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