tion of alignment, like that for orientation, is quite general. The argument applies, however, only when the light is incident from a restricted part of a sphere, for isotropic unpolarized light defines no axis with respect to which alignment can occur.

I wish to express my gratitude to Dr. R. H. Dicke, without whose inspiration and invaluable counsel this research could not have been undertaken or completed. I am also indebted to Dr. D. R. Hamilton for assistance in the early part of the work.

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Angular Correlation of Photons from Positron Annihilation in Light Metals*

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The angular correlation of gamma rays from positron annihilation in the light metals, Li, Be, Na, Mg, Al, K, and Ca, has been measured. The deviation of the two photons from colinearity is a measure of the total momentum of the annihilating electron-positron pair. If it is assumed that the positrons are thermalized before annihilation and that they annihilate only with electrons of a Sommerfeld free electron gas, the experimental results may be used to determine the Fermi energy of these electrons. It is interesting that the Fermi energies so determined are in good agreement with calculated values.

I. INTRODUCTION

THE possibility of determining electron velocities in metals from positron annihilation radiation was first pointed out by DuMond *et al.*[1] In their examination of the 0.511-Mev annihilation radiation these authors showed that the measured line width exceeded the instrumental width and ascribed the excess to motion of the electrons with which the positrons annihilate. Since that time DeBenedetti *et al.*[2] and other experimenters[3,4] have measured angular correlation of the two annihilation photons to determine the motion of the center of mass of the annihilating pair. The results of all these experimenters indicate that the center of mass of the annihilating pair has a momentum which is comparable with the momentum of conduction electrons in metals.

This paper reports an experiment in which the angular correlation of the annihilation photons was measured with slightly better angular resolution than had been previously used. Whereas other experimenters have attempted to relate the shape of the "tail" of the angular correlation curve to electron velocity, in this paper it is shown that the width of the observed curve is directly related to the momentum of the annihilating electrons.

Measurements in several metals have been made and the results are compared with theory.

II. EXPERIMENTAL

A schematic diagram of the apparatus is shown in Fig. 1. A source of annihilation radiation was placed at the middle of the horizontal line joining the two scintillation detectors. The rate of coincidences in the two detectors was measured as a function of the vertical displacement of the source. When corrected for background, this coincidence rate curve gives the angular correlation of the gamma-ray pairs.

The positron emitter was made by evaporating an aqueous solution of sodium chloride containing about 0.4 mC of Na^{22} on a thin (2 mg/cm²) aluminum foil. The approximately 0.5-in. diameter deposit and supporting foil were covered on both sides by another piece of the same foil. A sheet of the specimen metal

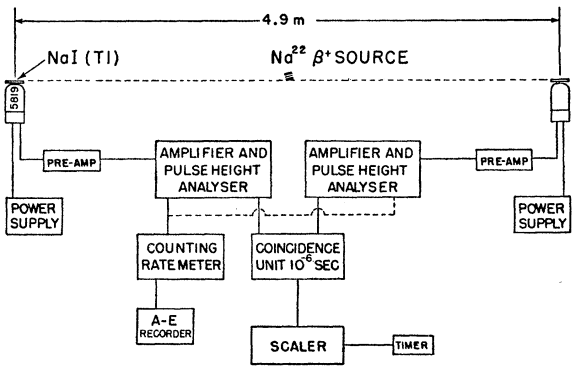

FIG. 1. Schematic diagram of apparatus.

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[1] DuMond, Lind, and Watson, Phys. Rev. **75**, 1226 (1949).

[2] DeBenedetti, Cowan, Konneker, and Primakoff, Phys. Rev. **77**, 205 (1950).

[3] P. E. Argyle and J. B. Warren, Can. J. Phys. **29**, 32 (1951), and J. B. Warren and G. M. Griffiths, Can. J. Phys. **29**, 325 (1951).

[4] H. Maier-Leibnitz, Z. Naturforsch. **6a**, 663 (1951).

was clamped to each side of the emitter, the resulting sandwich being the source of annihilation radiation. For measurements with the alkali metals and with some others, the emitter was protected by additional (5 mg/cm^2) aluminum foils. The flat, roughly circular, source was tipped to make an angle of about 10° with a line joining the detectors so that the apparent thickness (vertical dimension) of the source of annihilation radiation was independent of positron range in the metal specimen. As seen from the detectors, the source appeared from 3 to 4 mm thick.

The detectors were two NaI(Tl) crystal slabs approximately 2 in.$\times 1\frac{1}{2}$ in.$\times\frac{1}{4}$ in. mounted flat on RCA 5819 photomultiplier tubes. The 2 in.$\times\frac{1}{4}$ in. face was toward the source.

To eliminate pulses due to photons scattered in the crystal mount and photomultiplier tubes, the pulses from each detector were passed to a pulse height analyzer[5] set to select pulses of amplitude corresponding to a small energy range about 0.51 Mev. Coincidences within a resolving time of one microsecond between selected pulses from the two detectors were counted.

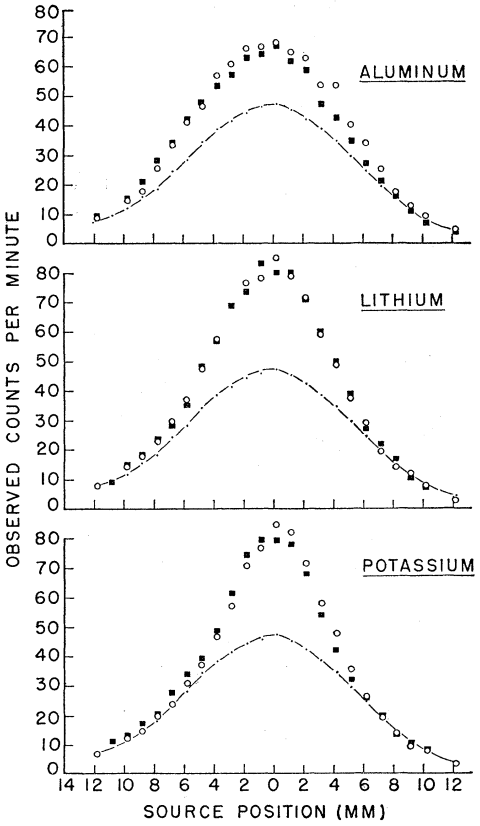

FIG. 2. Observed total angular correlation of annihilation photons from Al, Li, and K specimens clamped to the positron emitter. Separate runs are indicated by square and circular points. The dashed curve is the observed counting rate with no specimen metal against the emitter.

[5] Designed by R. E. Bell and R. L. Graham of Atomic Energy of Canada Ltd., Chalk River, Ontario.

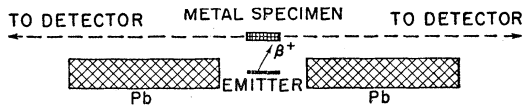

FIG. 3. Emitter and specimen arrangement for shielded emitter method.

The electronic circuits were operated from Thorensen regulators. In addition, the channel counting rates were maintained constant by manually adjusting the lower discriminator settings every fifteen minutes. This small regular readjustment corrected for gain variation in the photomultipliers and amplifiers and for the different gamma-ray attenuation in the various specimens. Because the channel counting rate varied rapidly with the lower discriminator setting, the average energy of the accepted pulses changed very little over the range of this adjustment.

The usual experimental procedure was to move the source vertically in 1-mm steps and to count for 20 minutes at each position. The maximum coincidence rate was about 100 cpm. Runs were made with various specimen metals in position and also with no specimens. These blank runs were used to determine the fraction of positrons stopping in the emitter.

In Fig. 2 are shown the observed angular correlations for some typical runs with potassium, lithium, and aluminum. The corresponding blank run is shown by the dashed line under each curve.

A second method of arranging the source was suggested by Dr. E. W. Guptill. In this method the annihilation radiation from the emitter is shielded from the detectors by blocks of lead (Fig. 3). Positrons from the emitter traverse about $\frac{1}{2}$ in. of air to reach the specimen which can be seen by the detectors. The emitter, shield, and specimen were raised and lowered as a unit.

Although this arrangement reduced the counting rate considerably, it had the advantage that the background was reduced by a much larger factor. In Fig. 4 are shown the results for calcium and beryllium from this type of experiment. The dashed line indicates the background, caused largely by scattered positrons.

III. THEORETICAL CALCULATION OF ANGULAR CORRELATION CURVES

To conserve momentum the angle between the two gamma rays from electron-positron annihilation must in general depart from 180° if the center of mass of the two particles was in motion. For low velocity the departure from 180° is $\phi = 2v/c$, where v is the transverse velocity of the center of mass of the particles and c is the velocity of light. It has been shown[6] that most positrons entering metals are thermalized in a time which is short compared with measured lifetimes. At thermal energies (1/40 ev) the positron velocity is very

[6] G. E. Lee-Whiting, Phys. Rev. **97**, 1557 (1955), and R. L. Garwin, Phys. Rev. **91**, 1571 (1953).

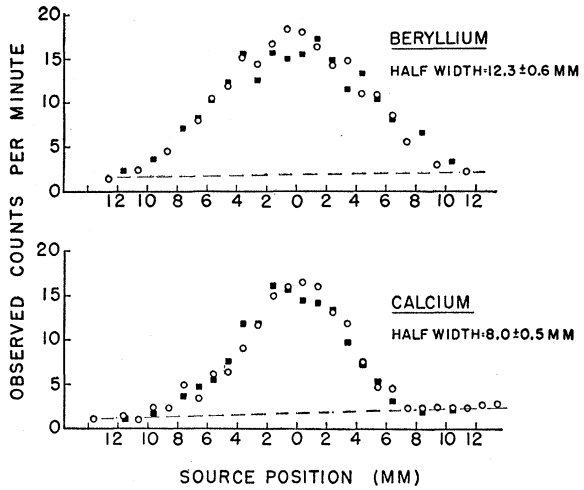

FIG. 4. Angular correlation of annihilation photons from Be and Ca observed by the shielded source method. The dashed curve is the background counting rate. Results of separate runs are indicated by round and square points.

small compared with conduction electron velocities, hence we can assume that the positron is at rest and that any motion of the center of mass of the annihilating pair is due to the velocity of the electron. Furthermore, DeBenedetti *et al.*[2] have pointed out that low-energy positrons would be expected to annihilate predominately with the conduction electrons: the Coulomb repulsion prevents the positrons from appreciably interacting with the inner electrons in the various elements.

In what follows we calculate the angular correlation which we expect to observe for positrons annihilating with the electrons in various metals assuming that the positrons are stationary and that the electrons have the momentum distribution predicted by the free electron theory.

1. Geometrical Resolution

The first step is to calculate the resolution function $R(z)$ due to finite thickness of source and detectors. (In the following analysis, the origin of the Cartesian coordinate system is at the mid-point of a line joining the two detectors; see Fig. 5. This line is the x-axis. The z-axis is vertical.) For a point source the resolution function, $R_0(z)$, is an isosceles triangle (Fig. 6) the width of the base of which is equal to the detector crystal thickness. The extension to a source of finite thickness, t, is merely the summation of $R_0(z)$ for

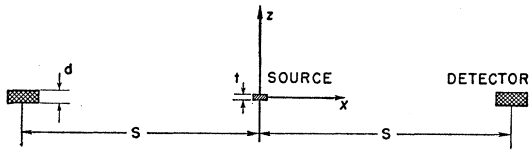

FIG. 5. Coordinate system with respect to source and detectors. For the arrangement used $d=7$ mm, $t=3$ mm, $s=2.45$ m.

various displacements of the point source. To simulate the actual source we assume that the activity is deposited uniformly over a $\frac{1}{2}$-in. disk and that the detectors see this disk at about 10° (see Sec. II). For these conditions, a plot of source strength, $I(z)$, against vertical distance is an ellipse as shown in Fig. 7. The resolution function may then be found by integrating $R_0(a)$ times $I(a-z)$ over the height of the source, i.e.,

$$R(z)=\int_{-t/2}^{+t/2} R_0(a)I(a-z)da.$$

$R(z)$ is shown in Fig. 6. The resolution function was also determined experimentally by moving the detectors so close to the source that the width caused by angular divergence of the gamma rays was easily allowed for by a small correction.

The detector and source widths introduce a horizontal resolution function similar to $R(z)$. However, an additional effect must be included. The measured efficiency of detection at the edges of the 2-in. crystals is about 0.7 of the efficiency in the center. The geometric resolution function corrected for counter efficiency gives an effective horizontal resolution function,

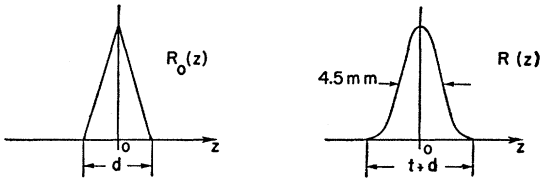

FIG. 6. Resolution functions $R_0(z)$ and $R(z)$ for a point source and a source of finite thickness.

$Y(y)$. As compared to a half-width (width at half-maximum) of 4.5 mm for $R(z)$, this function has a half-width of 29 mm.

2. Effect of Electron Motion

To calculate the effect of electron velocities we first recall that in the free electron theory of metals the tips of the momentum vectors of the conduction electrons uniformly fill a sphere. The volume of the sphere is proportional to the density of the free electrons in the metallic crystal.

If we consider annihilation radiation arising from electrons which have only one value, v_z, for the z component of velocity and v_y for the y component of velocity, we would observe experimentally a correlation having the shape of the $R(z)$ function times the $Y(y)$ function but shifted vertically from the origin an amount $sv_z/2c$ and horizontally an amount $sv_y/2c$, where s is the distance from source to detectors. In a free electron gas the number of electrons having a y and z component of velocity between v_y and v_y+dv_y and v_z and v_z+dv_z is given by

$$N(v_y,v_z)dv_ydv_z \propto (v_F^2-v_y^2-v_z^2)^{\frac{1}{2}}dv_ydv_z,$$

where v_F is the maximum velocity corresponding to the Fermi energy. To obtain the predicted curve for the conduction electrons we fold the RY product function with the electron distribution function, $N(v_y,v_z)$. The integrated function,

$$T(z,y)=\int_{-v_F}^{+v_F}\int_{-(v_F^2-v_z^2)^{\frac{1}{2}}}^{+(v_F^2-v_z^2)^{\frac{1}{2}}} N(v_z,v_y) \times R(z-sv_z/2c)Y(y-sv_y/2c)dv_ydv_z,$$

is proportional to rate of coincidences expected between the two detectors of Fig. 5 as the source of annihilation radiation is moved in the $y-z$ plane. Since we are interested in this curve for $y=0$ only we shall write simply $T(z)$. Curves calculated in this manner for various maximum electron velocities, corresponding to various metals, are shown in Fig. 8 and the half-widths are tabulated in Table I.

IV. ANALYSIS OF EXPERIMENTAL RESULTS

An observed angular correlation curve is the sum of curves representing annihilations which took place in the emitter and annihilations which occurred in the specimen. We assume that the area of the blank curve is proportional to the number of positrons that did not escape from the emitter and that the area of the curve observed with a specimen in position is proportional to the total number of positrons. Hence by subtraction of areas we can find the fraction of positrons which entered the specimen metal. The fraction which annihilated in the specimen is somewhat less than this because some of the positrons were scattered back to the emitter.

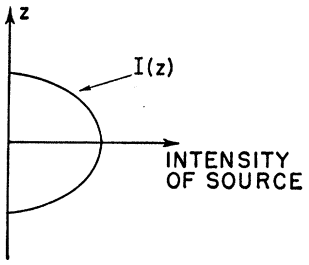

FIG. 7. Variation of source strength through the thickness of the source.

It was observed that the areas obtained from the many blank runs varied by about ±3 percent and that the areas obtained with the various specimens fluctuated about ±6 percent. As no definite cause could be assigned to this variation, which was a few times the statistical uncertainty, it was decided to take the average area of the blank curves to be proportional to the annihilation rate of positrons which do not leave the emitter. Similarly, the average area of the various specimen curves was taken to be proportional to the total annihilation rate. The specimen curves were then normalized to their average area before subtracting the curve representing annihilations in the emitter. For example, the procedure for an aluminum run was first to normalize by increasing the area by 100/92 (Table II). To determine the amount to be subtracted, we see from the table that about 64 percent of the annihilations occur in the emitter so that only about 36 percent of the positrons enter the specimen. Seliger[7] showed that a considerable fraction is reflected back to the emitter. After summing for repeated reflections, the net fraction returned to the emitter is found to be 0.22. Thus the fraction of positrons annihilating in the emitter when covered with an aluminum specimen is 64 percent plus 0.22×36 percent=72 percent=1.12×64 percent. Hence from the normalized aluminum curve we subtract 1.12 times the measured blank curve.

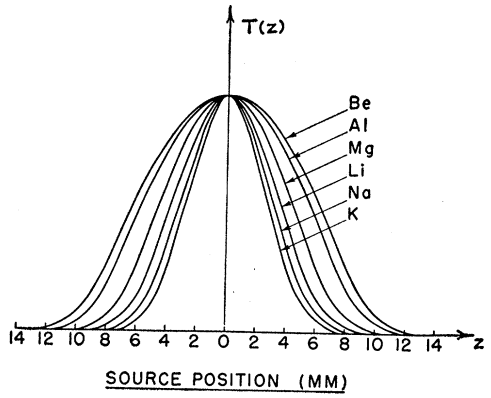

FIG. 8. Calculated angular correlation curves for various metals

Typical net curves are shown in Fig. 9. These curves may be compared with similar curves shown in Fig. 4 which were obtained by the shielded emitter method. The observed and calculated half-widths are listed in Table I.

In Fig. 10 is plotted the Fermi energy of the electrons of the specimen metal against the width of the correlation curve. The full line is the calculated curve and the points are observed values. The specimen was not tilted when used with the shielded emitter and hence

TABLE I. Calculated and observed half-widths of angular correlation curves for various metals. Resolution function, $R(z)$, half-width=4.5 mm.

	Free electrons per atom	Calculated[a] Fermi energy (ev)	Calculated half-width (mm)	Observed half-widths (mm) Subtracting blank method	Observed half-widths (mm) Shielded emitter method	Fermi energy from measured half-widths (ev)
K	1	2.04	6.0	6.0±0.2		2.1±0.3
Na	1	3.16	6.7	6.8±0.3		3.3±0.4
Li	1	4.75	7.8	7.7±0.3		4.6±0.5
Ca	2	4.72	7.8	7.7±0.3	8.0±0.5	4.8±0.4
Mg	2	7.16	9.3		9.4±0.3	7.4±0.5
Al	3	11.75	11.8	11.7±0.6	11.1±0.5	11.1±0.9
Be	2	14.3	12.9	12.8±0.6	12.3±0.6	13.6±0.9

[a] These Fermi energies have been calculated by assuming that the free electrons have the entire volume of the metal in which to move. If the ion core volume is excluded, the resultant Fermi energy will be higher by several percent for the heaviest of these metals.

[7] H. H. Seliger, Phys. Rev. 88, 408 (1952).

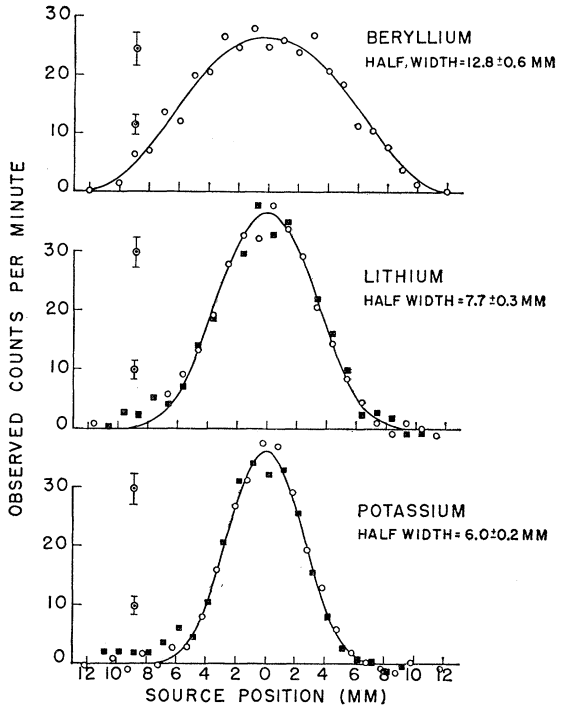

FIG. 9. Typical net curves resulting from subtraction of blank curve from the total coincident counting rate curves. The full line is the calculated curve normalized to equal area. The statistical uncertainty of the individual points is indicated by the vertical bars at the side of the curves.

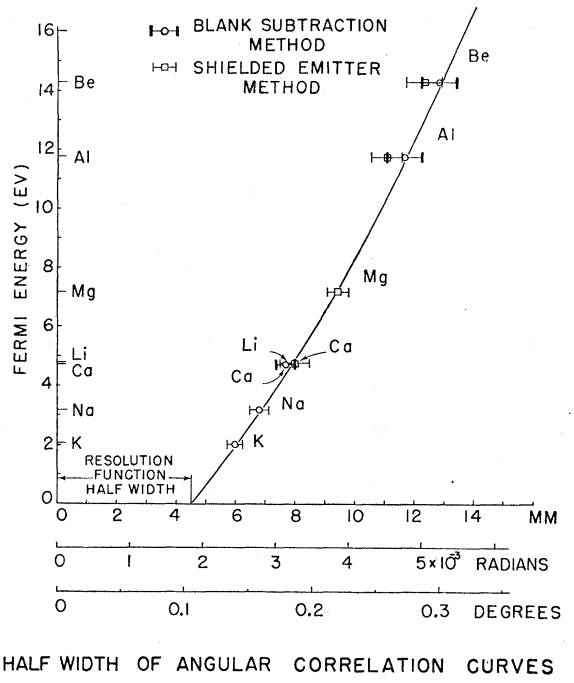

FIG. 10. Plot of Fermi energy against calculated half-width of correlation function. The measured half-widths are shown on the curve, the square and round points representing results of the shielded emitter and blank subtraction methods, respectively.

the curves calculated for this method are slightly narrower. However, the difference is within the experimental error, so that for clarity the calculated curve for this method has been omitted from the figure.

V. DISCUSSION

1. Experimental Uncertainties

On the assumption that the fluctuations in area have been due primarily to changes in coincidence efficiency, we have normalized all observed curves to the same area. Fortunately the widths of the net curves are not strongly dependent upon the method of normalizing. It can be shown for the narrow curves (K, Li) that the maximum change, 11 percent, in the area of the observed curve will cause only about a 4 percent change in the half-width of the net curve. The experimental error is also about 4 percent, and hence the mean uncertainty from the normalization is about half the statistical error shown in the table. For the wider curves the uncertainty will be less because both the total and blank curves have approximately the same width.

TABLE II. Typical areas of observed curves and effective positron reflection coefficients.

	Observed areas (relative values)	Net positron reflection coeff.	Percentage of positrons stopping in emitter
Observed area for Al	92	0.22	$64+36\times0.22=64\times1.12$
Observed area for Ca	110	0.26	$64+36\times0.26=64\times1.14$
Observed area for K	95	0.26	$64+36\times0.26=64\times1.14$
Average area of all runs	100		
Area of blank runs	64		
Percentage of positrons entering specimen	36		

The back-scattering was measured by Seliger[7] for positrons from a thin emitter whereas the positrons being reflected from the specimen in the present experiment have been degraded in energy by passing through the aluminum foil of the emitter. However, the error caused by this uncertainty cannot be very large because even a 50 percent change in the reflection coefficient will not change the half-width by more than the experimental error.

In the shielded emitter method, the background to be subtracted is in part caused by back-scattered positrons. Hence it is different for the various specimens and cannot be accurately determined merely by removing the specimen. The error assigned to these results contains an estimate of the background uncertainty.

Small-angle scattering of gamma rays (Rayleigh scattering) has been estimated and found to contribute negligibly to the broadening of the observed coincidence function.

2. Theoretical Assumptions and Implications

It is immediately evident from the experimental results that the two photons from electron positron

annihilation in light metals carry away an amount of momentum which is of the same order as the momentum of the conduction electrons. Since it has been shown that positrons thermalize very quickly,[6] we are led to believe that they annihilate chiefly with the conduction electrons of these metals.

If annihilations are assumed to occur with conduction electrons only, and to occur by a process which makes the total momentum of the two photons equal to the momentum of the annihilating electron, the quantitative experimental results may be interpreted in two ways: *Firstly*, we may assume that the momentum distribution of the conduction electrons is described by the free electron theory and deduce the velocity dependence of the annihilation cross section. The full line of Fig. 10 is calculated on this basis for a $1/v$ cross section. This corresponds to the assumption of equal probability of annihilation with all conduction electrons which was implicit in the arguments in Sec. III. If it were assumed that the annihilation cross section varies as $1/v^2$, the calculated half-widths would be too small by about three times the experimental error. Similarly, an annihilation cross section which was assumed to be independent of velocity would lead to calculated half-widths which are correspondingly large. *Secondly*, we may assume that the cross section varies as $1/v$ and use the experimental results to determine the Fermi energy of the conduction electrons. The Fermi energies so determined are listed in Table I. The results agree well with the values calculated from the free electron theory. The assumption of an annihilation cross section which varies other than as $1/v$ would yield Fermi energies which differ considerably from calculated values.

The mechanism of annihilation is not well understood. The most obvious process is the ordinary annihilation of an electron and positron in a collision, for which the cross section was first derived by Dirac.[8] However, as Bell and Graham have pointed out,[9] the cross section for direct annihilation leads to a lifetime several times that observed. In fact, for light metals, the electron density is not sufficient to account for the observed lifetime even if all the electrons are available for annihilations. It has been suggested[10] that Coulomb attraction may cause an increased electron density near the positron and hence a shorter lifetime. If this effect can be shown to shorten the calculated lifetime sufficiently, then the lifetime and angular correlation measurements will be consistent. If not, it may be necessary to consider the formation of positronium in metals in a time short compared with $\sim 10^{-10}$ sec, the observed lifetime. The atom might then annihilate in a time of the order of the singlet lifetime, 1.25×10^{-10} sec. This picture is not free from obstacles. It would be necessary to have a mode of formation in which the positronium atom retains nearly all of the conduction electron momentum and in which therefore, a photon or possibly an Auger electron removes the excess energy. In addition, the atom must retain its kinetic energy during collisions in the metal. Other pictures of varying complexity are also possible. The unsolved problem remains: to account for the observation that approximately 10^{-10} sec after a fast positron enters a metal, annihilation photons emerge with a total momentum which appears to be that of a conduction electron.

[8] P. A. M. Dirac, Proc. Cambridge Phil. Soc. **26**, 361 (1930).
[9] R. E. Bell and R. L. Graham, Phys. Rev. **90**, 644 (1953).
[10] Reference 2, footnote 18.

ACKNOWLEDGMENTS

The determination of electron velocities by measuring the width of the angular distribution curve instead of the "tail," as had been previously attempted, was suggested to us by Dr. G. E. Lee-Whiting to whom we are indebted for many helpful discussions. It is also a pleasure to acknowledge the advice and assistance of Dr. R. L. Graham and Mr. R. M. MacIntyre who contributed to the preliminary experiments. Many others of the Physics Department of Dalhousie University and the Physics Division of Atomic Energy of Canada Limited have assisted through discussions.

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