MOMENTUM DISTRIBUTION OF METALLIC ELECTRONS BY POSITRON ANNIHILATION[1]

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ABSTRACT

The angular correlation of photons from the two-photon decay of positrons has been measured for positrons annihilating in some 34 elements, mostly metals. These data give the momentum distribution of photons and hence of the center of mass of the annihilating electron–positron pairs. The momentum distributions are discussed in terms of the velocity dependence of the annihilation probability. It is concluded that the observed momentum distributions are primarily the momentum distributions of the conduction electrons in the metals. A higher momentum component is observed, which is attributed to ion core effects.

INTRODUCTION

The annihilation of free electron–positron pairs usually occurs by the emission of two photons, which, to conserve momentum, are emitted at 180° to each other in the center of mass of the annihilating particles. If the center of mass is in motion with respect to an observer the angle between the photon directions departs from 180° by an amount of the order of $v_{c.m.}/c$, where $v_{c.m.}$ is the velocity of the center of mass and c is the velocity of light. For the low velocities of interest here the departure of the angle between the photon directions from 180° is proportional to the component of momentum of the annihilating pair which is parallel to the bisector of the propagation directions.

The first measurements of the angular correlation of annihilation photons were made by Beringer and Montgomery in 1942. Following this Vlasov and Tsirelson (1948) showed that 95% of the photon pairs were emitted within 180°±1°, from which they concluded that the kinetic energies of most of the annihilating positrons were less than 80 ev. Soon after these experiments Dumond *et al.* (Dumond, Lind, and Watson 1949) showed that the line width of the (0.511 Mev.) annihilation radiation was slightly greater than their instrument resolution. The excess width could be accounted for by assuming that the electron or positron had a kinetic energy at annihilation of 10–20 ev. Following this, DeBenedetti *et al.* (DeBenedetti, Cowan, and Konneker 1949; DeBenedetti, Cowan, Konneker, and Primakoff 1950) arrived at a similar conclusion from their study of the angular correlation of the annihilation radiation. Warren and co-workers (Argyle and Warren 1951; Warren and Griffiths 1951; and Erdman 1955) also measured the angular correlation of photons from annihilations in various elements and compounds.

Maier-Leibnitz' measurements (1951) of the angular correlation of the radiation from positrons annihilating in several metals showed clearly that there were differences between various metals and that the mean momentum of the annihilating pairs in some heavy metals was greater than the momentum of conduction electrons.

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In 1955 some measurements were made by Green and Stewart (1955, and Stewart and Green 1954) which showed that the half-width of the angular distribution was a good measure of the Fermi energy of light metals. Other experimenters (Page, Heinberg, Wallace, and Trout 1955; Page and Heinberg 1956) have shown that angular correlation measurements give information about the mechanism of positron annihilation in solids and in some cases about the electrons of the solid with which the positrons annihilate. Extensive work with metals done at Professor DeBenedetti's Laboratory (Lang, DeBenedetti, and Smoluchowski 1955) has given results similar to and in agreement with those presented in this paper.

Here we present* measurements of the angular distribution of photons from positron annihilation in 30 metals and four other closely related elements. It is shown that a simple transform of the angular distributions can yield the distribution in momentum of the annihilating electron–positron pairs. These derived distributions are presented for all the specimens and are discussed in terms of the momentum distribution of free electrons, which they often closely resemble.

EXPERIMENTAL

A schematic diagram of the apparatus is shown in Fig. 1. Radiation from annihilation of positrons in the specimen was detected in two 2 in. × 2 in.

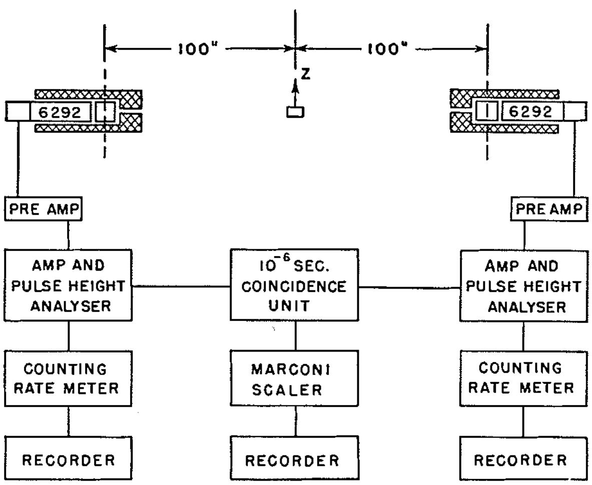

FIG. 1. Schematic diagram of the apparatus.

cylindrical NaI(Tl) crystals and RCA 6292 photomultipliers mounted behind 0.050 in. × 1.5 in. horizontal slits in lead shields. The rate of coincidences in the two detectors was measured as a function of the vertical displacement of the specimen from the mid-point between the counters. Automatic equipment moved the specimen at 5 minute intervals and recorded the counting rate. The pulse height analyzers selected pulses corresponding to photons in the energy range from about 200 to 600 kev.

*A preliminary report of this work was given in a letter to *The Physical Review* (Stewart 1955).

The positron emitter used was Cu^{64} (12.8 hr.) made by irradiating copper foils in the NRX reactor. Two methods of bombarding the approx. $\frac{5}{8}$ in. diam. specimen with positrons were used. In one, a multilayer sandwich was made from thin foils of the specimen metal alternating with approximately 1 mg./cm.^{2} copper leaf. The total thickness and vertical dimension of such a sandwich was usually about 0.06 in. With this technique only about 3% of the positrons annihilated in the copper. Because the shape and width of the angular correlation curve for positrons annihilating in copper were not very different from the results obtained for most of the metals examined by this method, no corrections were made for annihilations in the copper leaf.

In the other method a much thicker piece of copper foil was used as a positron emitter. It was mounted about $\frac{1}{2}$ in. from the specimen and was shielded by several inches of lead from the direct view of the detectors. This method was used for many specimens, including the chemically active metals, which were supported in the center of a vacuum chamber. Under these circumstances positrons entered from above through a thin mica window.

RESULTS

The angular correlation measurements for 34 specimens are shown in Fig. 2. The data shown for each element are the sum of many runs through the angular range indicated. A correction for source decay has been made. The total number of counts at the peak position is greater than 6000 for all except some heavy metals and is at least twice this value for most of the alkali metals.

Before the momentum distributions of annihilation photons are obtained from the angular distributions, two instrumental effects must be considered. The more important is the combined effect of detector slit width and specimen thickness. This produces an approximately Gaussian shaped angular resolution function of half-width about 0.6×10^{-3} radians for most of the runs, increasing to about 2×10^{-3} radians for some light metals. A correction for this instrumental resolution width was calculated by the parabolic method of Eckart (1937) and was applied to the observational points. The effect of the correction is to increase the height of the peak and to sharpen the corners of the angular correlation curves. The maximum correction was applied to measurements with the Mg specimen and amounted to only 4% at the peak.

The other is the effect of the finite length (1.5 in.) of the slits, which causes a loss in counting efficiency for photon pairs with large angular deviation from 180°. In terms of the total momentum of the annihilating particles the decrease in detection efficiency is found to be about 6% and 25% for total momenta 3 and 6 (times $2mc\times10^{-3}$) respectively. A correction has not been made for this effect because it is well within the experimental errors in the lower momentum region of interest.

The momentum distribution of the annihilation photons $N(k)$, and the corresponding momentum space density $\rho(k)$, were obtained using the relations

$$N(k) \propto dI/dz\cdot z, \qquad \rho(k) \propto dI/dz\cdot 1/z,$$

which are derived in the Appendix. In these expressions I is the coincidence counting rate as a function of z, the vertical displacement of the specimen

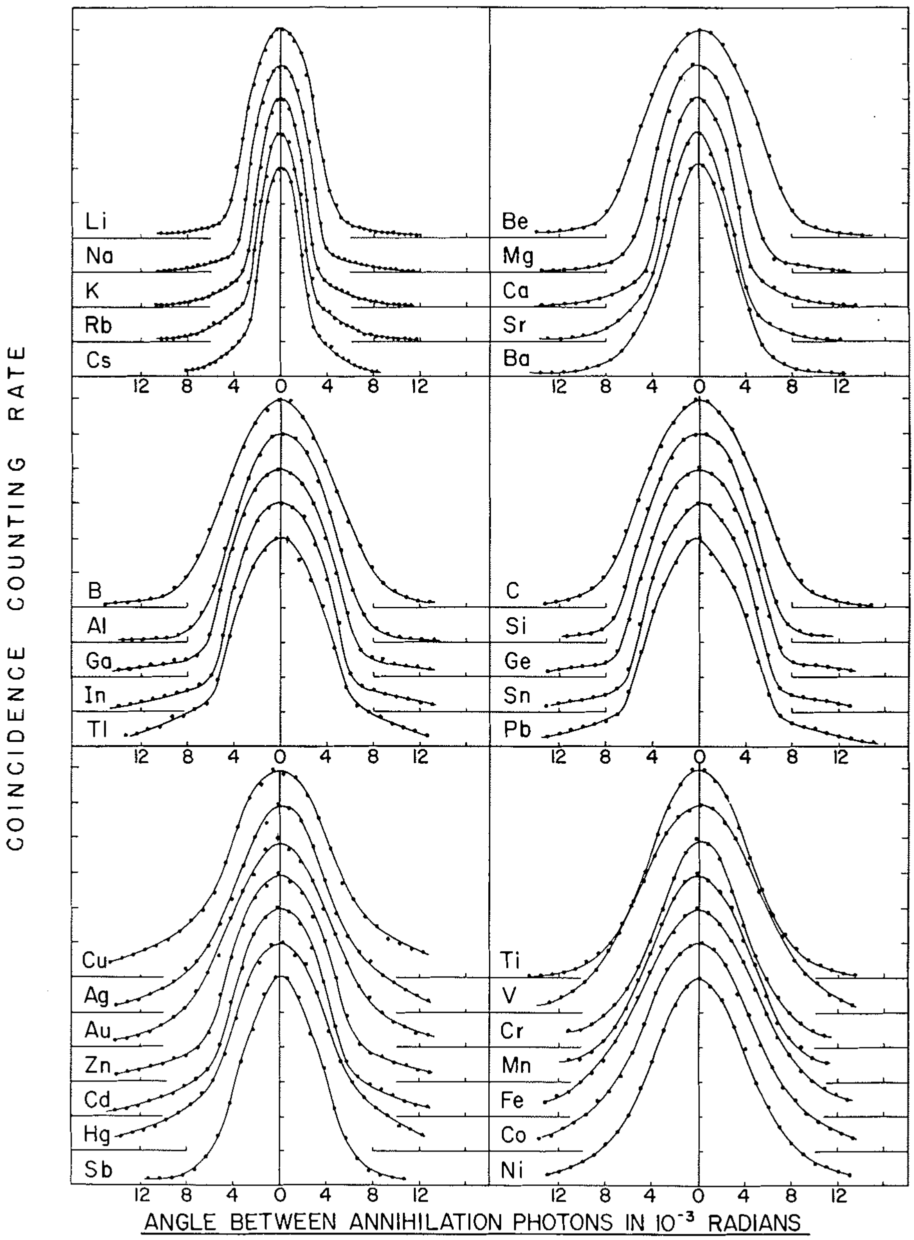

FIG. 2. Observed angular correlation of photons from positrons annihilating in various elements. The data have been normalized to make all curves the same height.

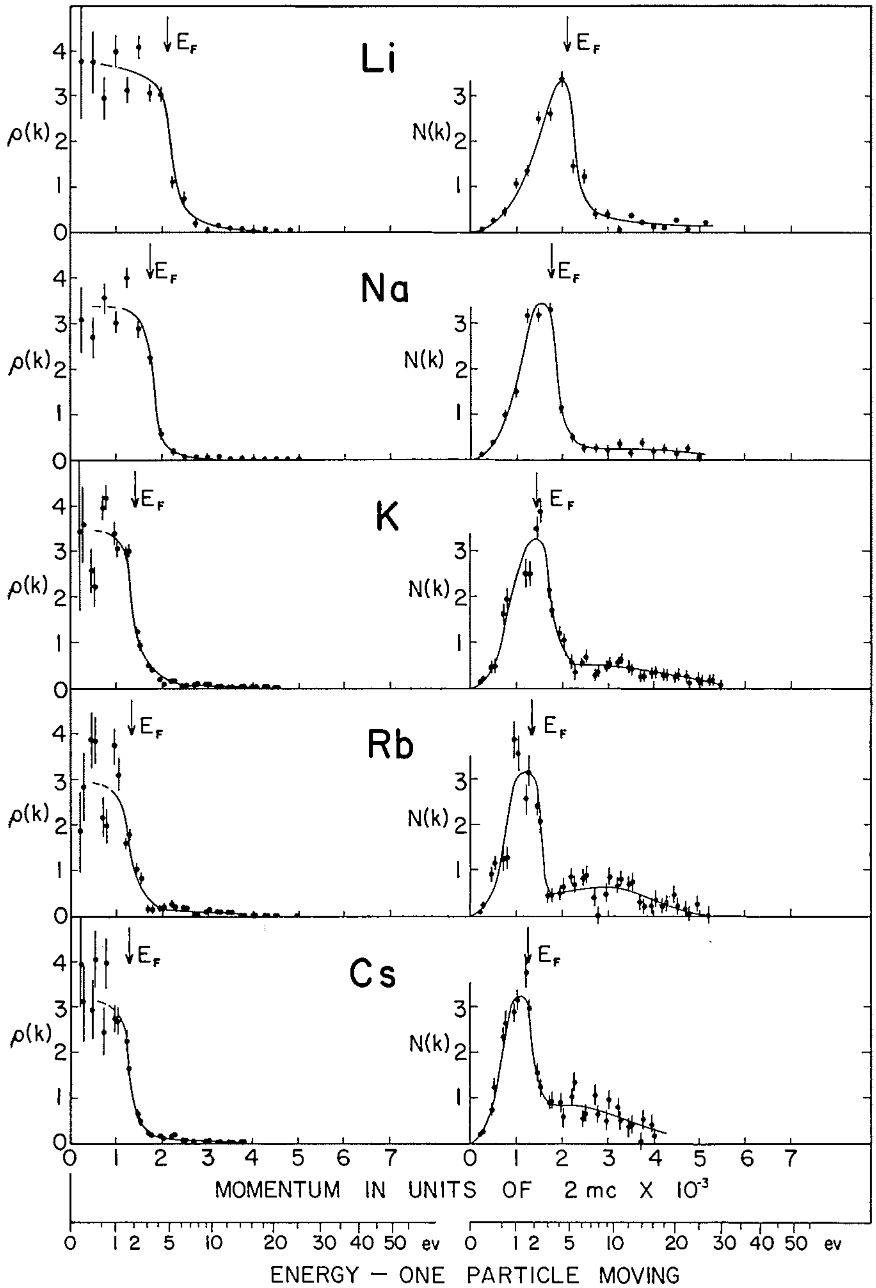

FIG. 3. Momentum distributions, $N(k)$, and momentum space densities, $\rho(k)$, of photons from positron annihilations in the alkali metals Li, Na, K, Rb, and Cs of Group Ia of the Periodic Table.

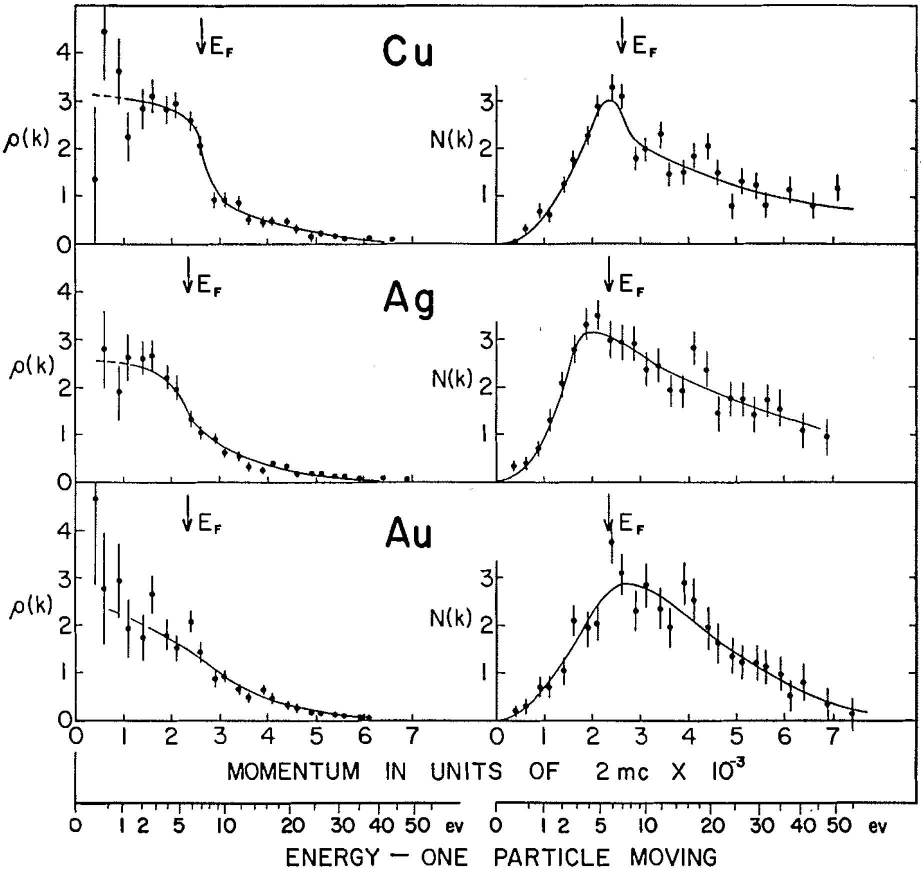

FIG. 4. Momentum distributions, $N(k)$, and momentum space densities, $\rho(k)$, of photons from positron annihilations in the noble metals Cu, Ag, and Au of Group Ib.

from the line joining the detector slits. The centers of the observed angular distributions were taken to be $z = 0$. The value of the chord $(I_1-I_2)/(z_1-z_2)$ was used for the derivative dI/dz at $z = (z_1+z_2)/2$, where I_1 and I_2 are the experimentally observed counting rates (with above correction) at z_1 and z_2 respectively. The momentum distribution and momentum space density so calculated are shown in Figs. 3 to 11. The errors indicated in the figures are the statistical errors of counting and an estimate of the effect of uncertainty in z. The full line has been arbitrarily drawn through the points. The arrows marked E_F indicate the momentum of an electron at the Fermi surface* for 1, 2, 3, and 4 electrons per atom in metals of Groups I, II, III, and IV respectively. In Fig. 11 (Fe, Co, and Ni) two arrows are shown for 0.5 and 1 electron per atom. The energy scale on each figure was calculated on the assumption that the momentum is due to the motion of one of the annihilating particles only.

*The Fermi energy was computed from the usual formula given for example in Mott and Jones, *Properties of Metals and Alloys*, Chap. II, Eq. 19.

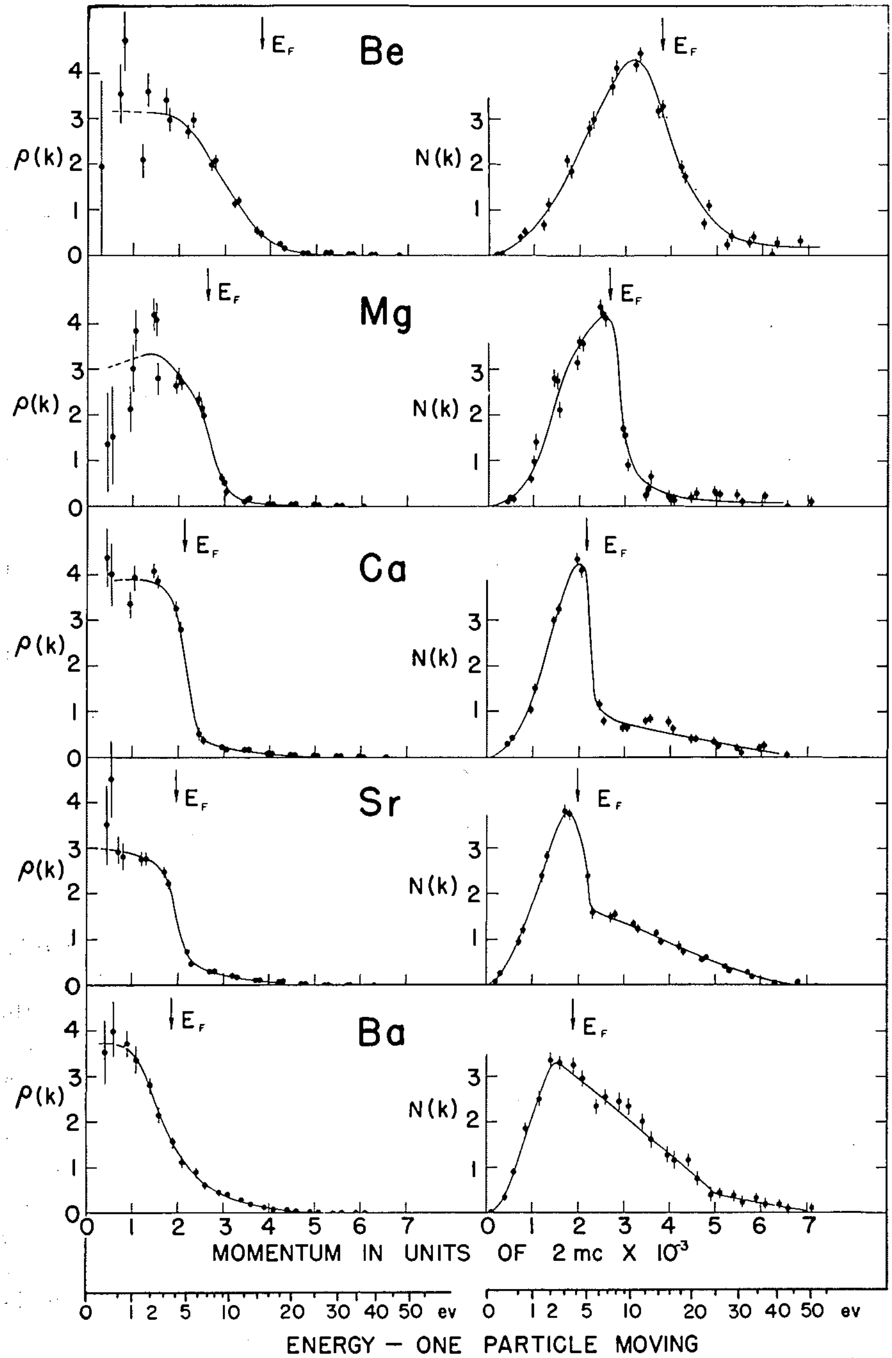

FIG. 5. Momentum distributions, $N(k)$, and momentum space densities, $\rho(k)$, of photons from positron annihilations in the alkali earth metals Be, Mg, Ca, Sr, and Ba of Group IIa.

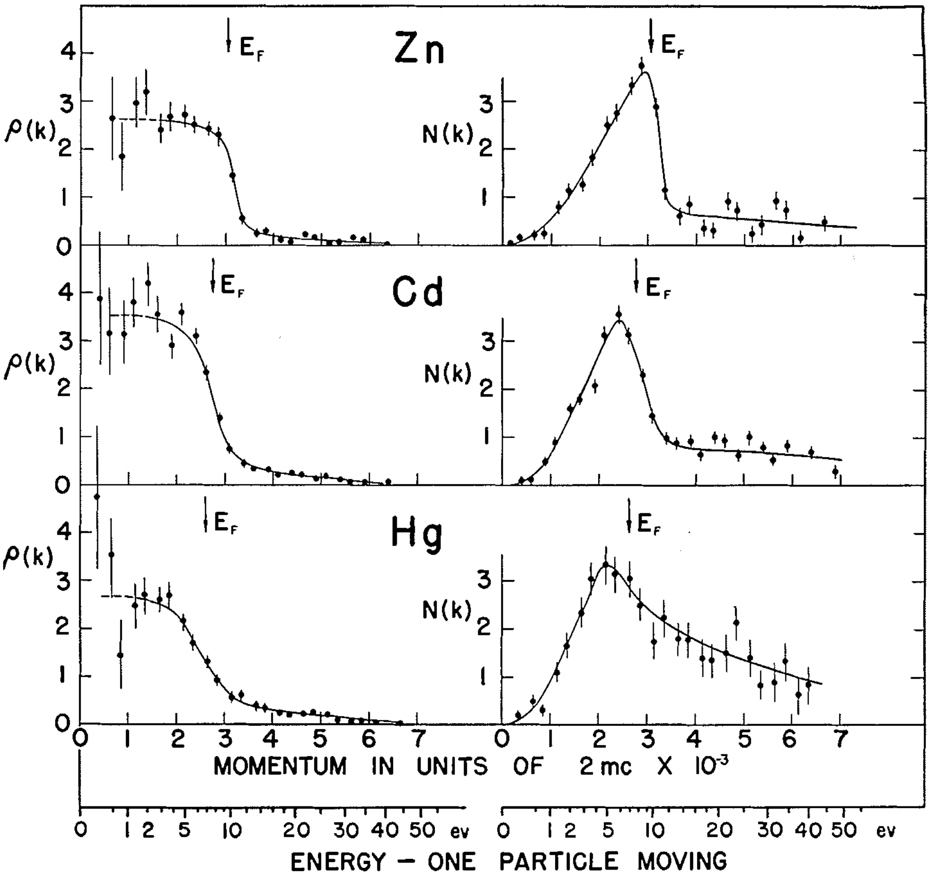

FIG. 6. Momentum distributions, $N(k)$, and momentum space densities, $\rho(k)$, of photons from positron annihilations in metals Zn, Cd, and Hg of Group IIb.

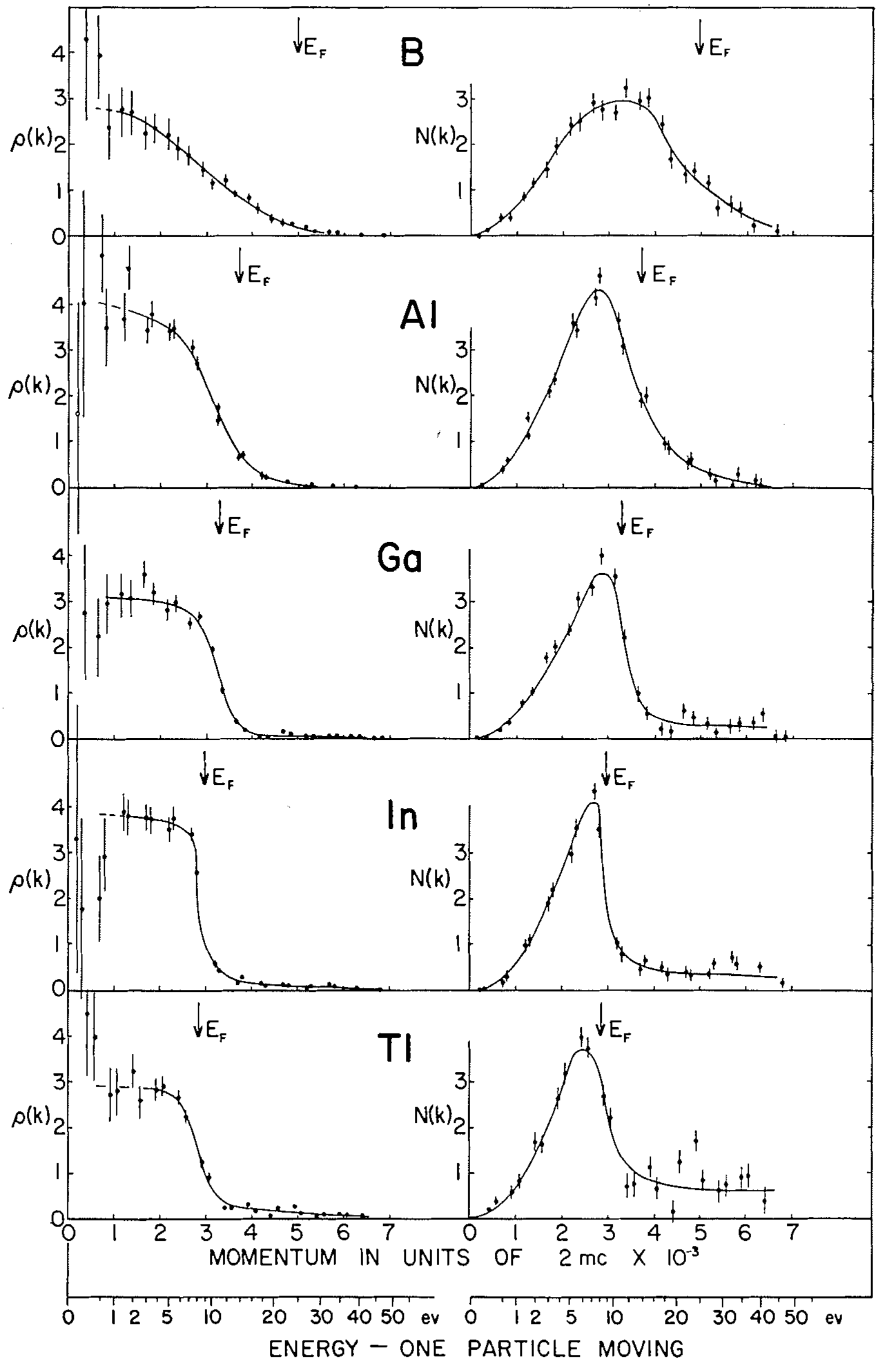

FIG. 7. Momentum distributions, $N(k)$, and momentum space densities, $\rho(k)$, of photons from positron annihilations in the metals B, Al, Ga, In, and Tl of Group III.

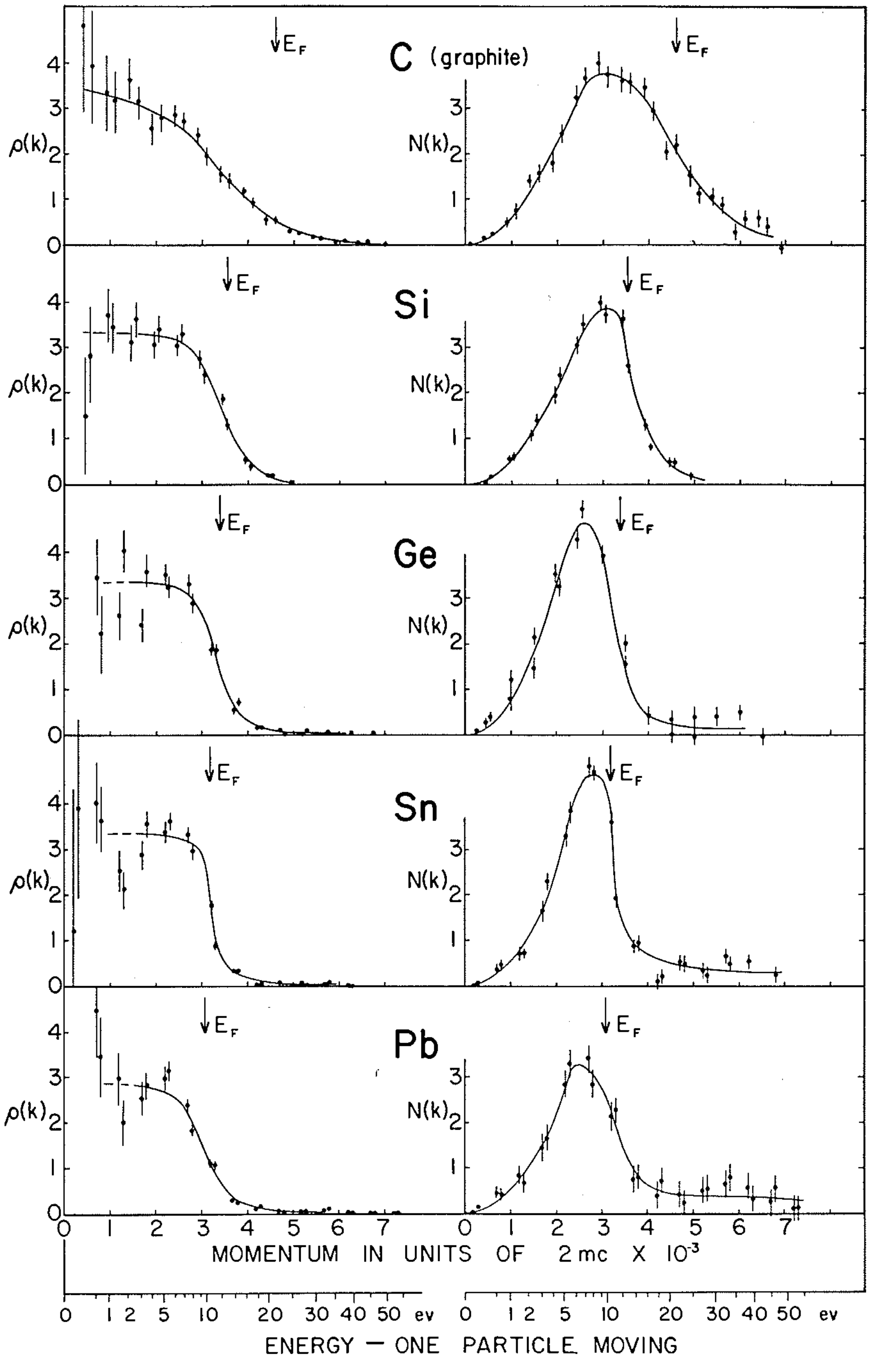

FIG. 8. Momentum distributions, $N(k)$, and momentum space densities, $\rho(k)$, of photons from positron annihilations in the elements C, Si, Ge, Sn, and Pb of Group IV.

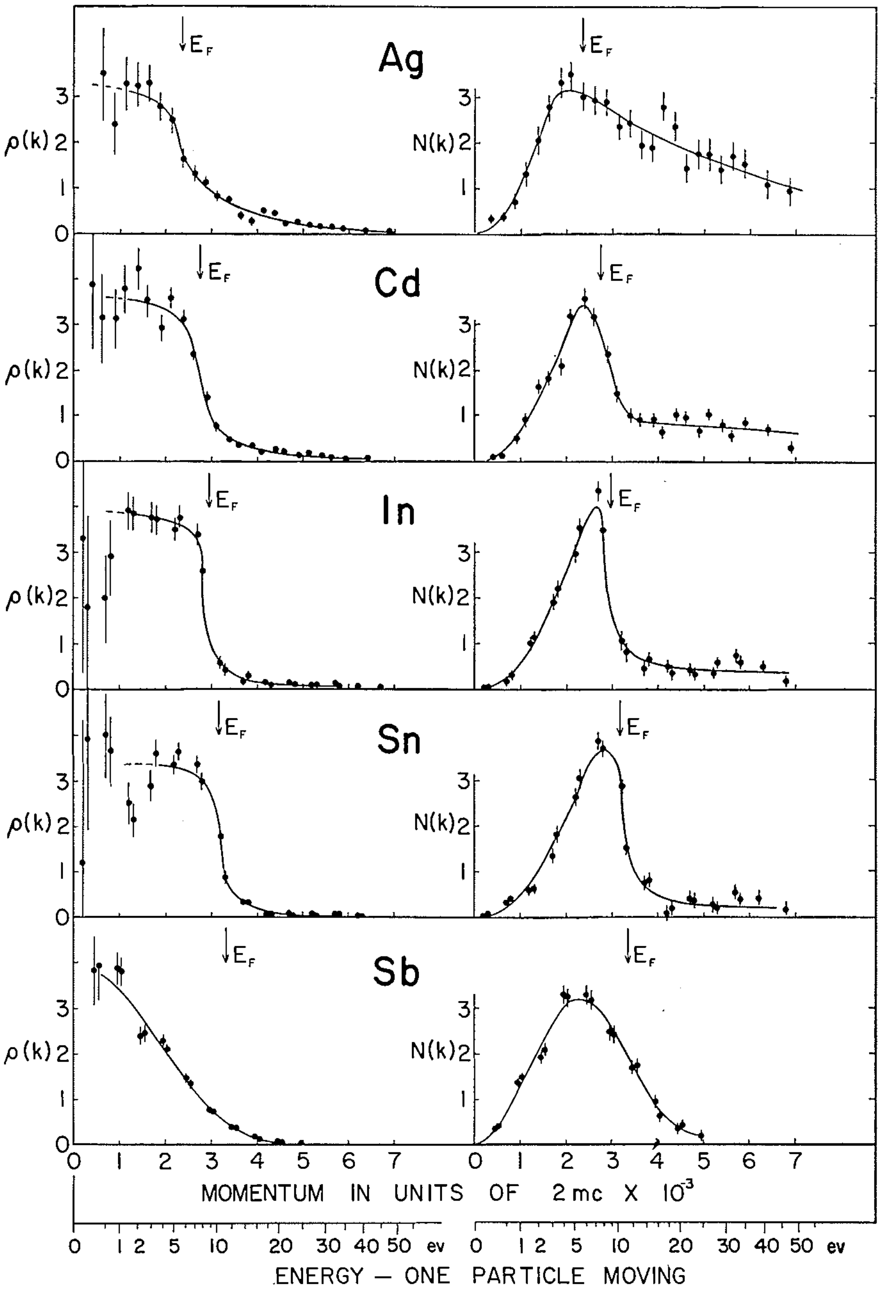

FIG. 9. Momentum distributions, $N(k)$, and momentum space densities, $\rho(k)$, of photons from positron annihilations in the metals Ag, Cd, In, Sn, and Sb of Period IVb.

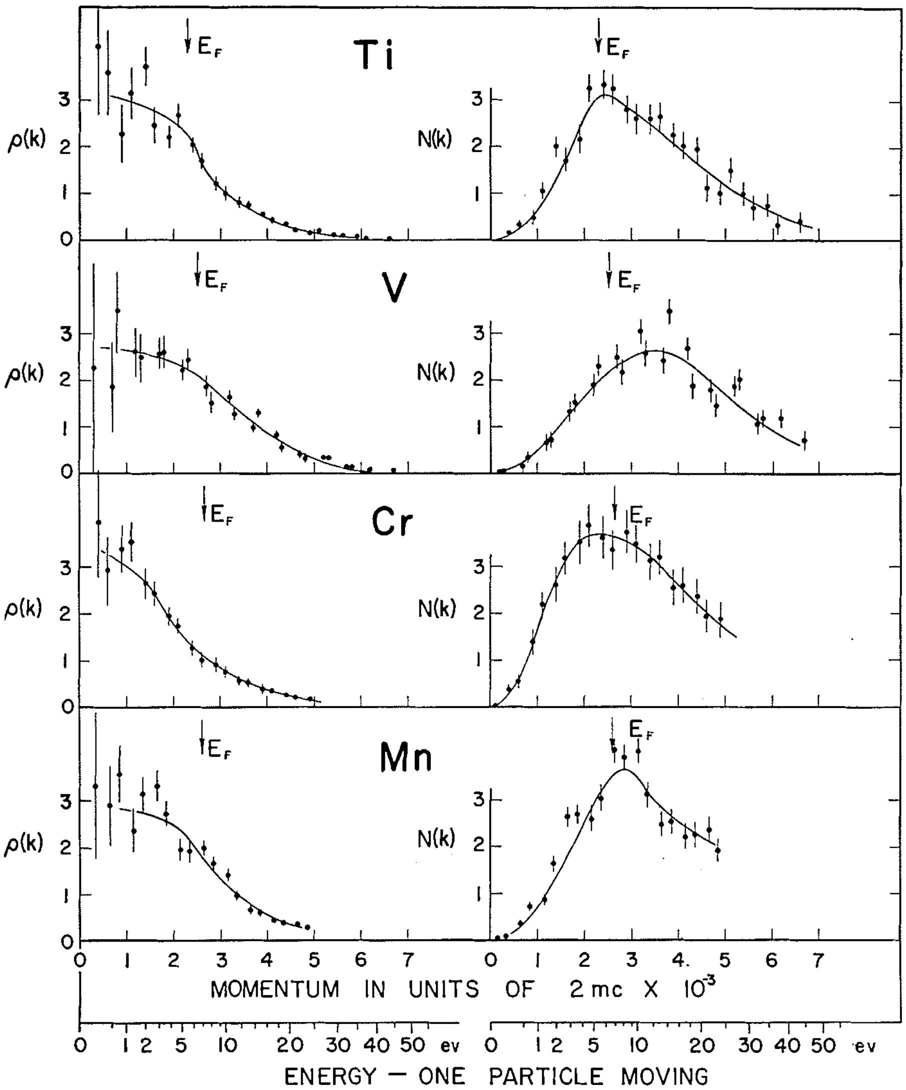

FIG. 10. Momentum distributions, $N(k)$, and momentum space densities, $\rho(k)$, of photons from positron annihilations in the transition metals Ti, V, Cr, and Mn.

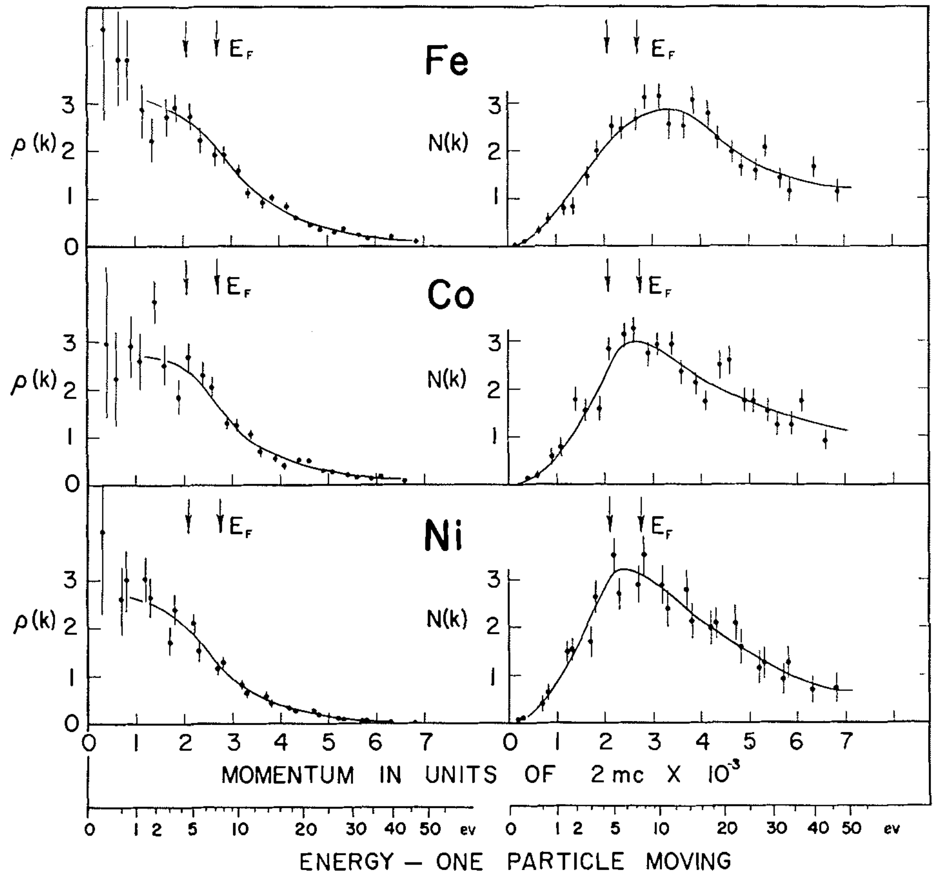

FIG. 11. Momentum distributions, $N(k)$, and momentum space densities, $\rho(k)$, of photons from positron annihilations in the transition metals Fe, Co, and Ni.

DISCUSSION

Momentum Distributions

The momentum distributions of annihilating electron–positron pairs appear to have two components, which are clearly seen in some of the alkali and alkali earth metals. The similarity between the lower momentum components of the momentum distributions obtained in this experiment and the momentum distributions expected for conduction electrons of the free electron metals, i.e. the alkali metals, is striking. (It should be remembered that $\rho(k)$ and $N(k)$ for electrons of a free electron gas are constant and quadratic respectively out to the Fermi cutoff, where they fall to zero.) That the maximum momentum cutoff occurs at the calculated Fermi limit provides experimental support for Lee-Whiting's (1955) calculation which showed that positrons in metals thermalize in a time short compared with annihilation lifetime. Thus the total momentum of the photons is very likely the momentum of the annihilation electron. The form of the momentum distribution below the cutoff is somewhat similar to the free electron theory momentum distribution and implies that the annihilation probability is not strongly velocity dependent. For example, an annihilation probability of the form $1/v$, which is predicted by a coulomb interaction between a free electron and positron, is precluded. The almost velocity independent annihilation probability calculated by Ferrell (1956) is in agreement with these results. For metals other than the alkalies the momentum distribution and the maximum momentum cutoff are not expected to resemble so closely the free electron theory but will contain lattice effects. For example the two-electron alkali earth metals have Fermi surfaces near the reciprocal lattice zone boundaries and thus the electron momentum near the Fermi limit is reduced below the free electron theory value, even being zero at some symmetry points on the faces of the zone. The result of lattice effects will be to change the quadratic function $N(k)$ of the free electron theory by reducing the number of high momentum electrons and increasing the number with lower momentum. For the three-electron Al structure the Fermi surface in most places extends past the first zone boundaries and in some places probably past the free electron Fermi sphere. It can be seen that a small number of electrons in Al have momenta greater than the Fermi limit, as indeed is the case for several others.

Higher Momentum Component

The higher momentum component of the distribution observed in this experiment is probably due to an effect of the ion cores. At first sight it might appear that this component is caused by positrons annihilating with electrons bound to the ion core, because the importance of the effect increases roughly as the volume of the core. However another explanation is also possible. DeBenedetti *et al.* (1950) showed that because the positron space wave function is excluded (by coulomb repulsion) from the interior of the ion cores, the momentum wave function is many valued having momenta which correspond to the wave numbers of the points of the reciprocal lattice. In this explanation too, the relative importance of the higher momentum components of the positron wave function increases with increase in core volume. Ferrell (1956) has shown that this excluded volume effect is probably the more important.

Lifetime

The rate of annihilation of positrons in metals has been discussed many times.* The measured lifetime is about $1.5\pm0.7\times10^{-10}$ sec.† whereas the expected lifetime for a non-coulomb interacting positron in a sea of electrons of density similar to the density of conduction electrons in metals is one or two orders of magnitude longer and varies with electron density. Inclusion of the coulomb interaction makes the calculation of annihilation probability difficult. The simplest improvement, the introduction of a coulomb interaction between the otherwise free annihilating pairs, increases the rate of annihilation almost enough but gives a $1/v$ annihilation probability, which, as discussed

*See for example the review article by Ferrell (1956).

†Recent experiments of Gerholm (1956) and Minton (1954) yield lifetimes of about 2.5×10^{-10} sec. which are not in agreement with most other measurements.

above, seems unlikely in view of the results of the present experiment. The more complicated case of allowing coulomb interaction between all particles and not only the annihilating pair has been discussed by Ferrell (1956). His calculations yield an annihilation probability which is approximately independent of velocity and give a lifetime in agreement with observation for the metals of higher conduction electron density (e.g. Al) but too low by a factor of about four for lower density metals (e.g. Na). As he points out, however, one would not expect the lifetime of an interacting positron in an electron sea to exceed the lifetime of the positronium ion, $e^-e^-e^+$, which is the bound (0.2 ev.) system of particles with the greatest density of electrons at the positron. The electron density in this system is about 50% greater than in the positronium atom, e^-e^+. The annihilation rates of the singlet and triplet positronium atom are 8.0×10^9/sec. and 7.1×10^6/sec. respectively (Ore and Powell 1949). The annihilation rate of the ion is then $1.5(\frac{1}{4}\times8.0\times10^9+\frac{3}{4}\times 7.1\times10^6)$/sec. $= 3.0\times10^9$/sec. This rate corresponds to a lifetime of 3.4×10^{-10} sec., which is somewhat greater than the measured lifetime of positrons in metals. If one considers a positron located in a lattice defect or an interstitial positron in a crystal it is reasonable to expect that the wave functions of the valence band electrons are not much altered by its presence. The wave functions will be slightly increased at the positron and virtually unchanged in the remainder of the crystal. The sum of these small increases in each electron wave function may be sufficient to concentrate at the positron a negative charge density equivalent to that in the positronium ion. Hence we might expect the positron to have a short lifetime and to annihilate with an electron which has a momentum almost exactly that of the unperturbed valence electron. The observed momentum distributions are in agreement with this simple picture of positron annihilation in metals.

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APPENDIX

Proportionality Constant between z and k_z

We define **k** to be the momentum of the center of mass of the electron–positron pair at the time of annihilation. If k_z is the z component of this momentum, a coincidence between the annihilation photons can be detected only when the source is displaced from a line joining the counter slits by an amount $z = k_z d/2mc$, where $2d$ is the distance between the counters, m is the electron mass, and c the velocity of light.

Derivation of Differentiation Relation

When the source is displaced an amount z the coincident counting rate $I(z)$ is proportional to the total number of pairs with $k_z = 2mc\, z/d$, that is

$$I(z) = \text{const.} \iint \rho(\mathbf{k})\, dk_x\, dk_y,$$

where $\rho(\mathbf{k})$ is the momentum space density of annihilating electron–positron pairs and the integral is taken over the plane of constant k_z. In this expression we assume that variations in $\rho(\mathbf{k})$ are small within a region Δk_z corresponding to the slit width, and that the length of the slits is much greater than the extent of $\rho(\mathbf{k})$, i.e. slit length $\gg k_{\max}d/2mc$. As discussed above small corrections for the finite slit width and slit length have been calculated and the former applied. If the distribution in momentum space is assumed isotropic and denoted by $\rho(k)$, the integral becomes

$$I(z) = \text{const.}\, 2\pi \int_{k_z}^{\infty} k\rho(k)\, dk,$$

from which we have by differentiating with respect to k_z

$$\rho(k) = \frac{-(d/2mc)^2}{2\pi\, \text{const.}} \frac{dI}{dz} \frac{1}{z},$$

and for momentum distribution

$$N(k) = 4\pi k^2 \rho(k)$$
$$= \frac{-2}{\text{const.}} \frac{dI}{dz} z.$$

REFERENCES

ARGYLE, P. E. and WARREN, J. B. 1951. Can. J. Phys. **29**, 32.
BERINGER, R. and MONTGOMERY, C. G. 1942. Phys. Rev. **61**, 222.
DEBENEDETTI, S., COWAN, C. E., and KONNEKER, W. R. 1949. Phys. Rev. **76**, 440(L).
DEBENEDETTI, S., COWAN, C. E., KONNEKER, W. R., and PRIMAKOFF, H. 1950. Phys. Rev. **77**, 205.
DUMOND, J. W. M., LIND, D. A., and WATSON, B. B. 1949. Phys. Rev. **75**, 1226.
ECKART, C. 1937. Phys. Rev. **51**, 735.
ERDMAN, K. L. 1955. Proc. Phys. Soc. **68**, 304.
FERRELL, R. A. 1956. Revs. Mod. Phys. **28**, 308.
GERHOLM, T. R. 1956. Arkiv Fysik, **10**, 523.
GREEN, R. E. and STEWART, A. T. 1955. Phys. Rev. **98**, 486.
LANG, G., DEBENEDETTI, S., and SMOLUCHOWSKI, R. 1955. Phys. Rev. **99**, 596(L).
LEE-WHITING, G. E. 1955. Phys. Rev. **97**, 1557.
MAIER-LEIBNITZ, H. 1951. Z. Naturforsch. **6a**, 663.
MINTON, G. E. 1954. Phys. Rev. **94**, 758.
ORE, A. and POWELL, J. L. 1949. Phys. Rev. **75**, 1696.
PAGE, L. A. and HEINBERG, M. 1956. Phys. Rev. **102**, 1545.
PAGE, L. A., HEINBERG, M., WALLACE, J., and TROUT, T. 1955. Phys. Rev. **98**, 206(L).
STEWART, A. T. 1955. Phys. Rev. **99**, 594(L).
STEWART, A. T. and GREEN, R. E. 1954. Bull. Am. Phys. Soc. **29**, No. 7, H12 (Phys. Rev. **98**, 232(A) (1955)).
VLASOV, N. A. and TSIRELSON, E. A. 1948. Doklady Akad. Nauk S.S.S.R. **59**, 879.
WARREN, J. B. and GRIFFITHS, G. M. 1951. Can. J. Phys. **29**, 325.