

## On the Angular Distribution of Two-Photon Annihilation Radiation<sup>1</sup>

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An experimental determination, using anthracene scintillation counters in coincidence, has been made of the departure from antiparallelism of the two photons in the pair annihilation process in an Au absorber. The observed mean angular departure,  $\approx 1/137$  radians, arises from the motion of the centers of mass of the annihilating pairs; the mean momentum of these is found from the experimental data to be equal to  $1.2 mc/137$ , a value to be compared with a lower limit of  $0.8 mc/137$  obtained from an included theoretical discussion in which the momentum probability am-

plitudes of the annihilating electrons and positrons (in the absorbing metal) are estimated and appropriately combined. In these estimates it is shown that the positrons become thermalized by collisions with the vibrating lattice atoms in times considerably shorter than the annihilation time, so that only their zero point motions, together with the zero point motions of the annihilating (valence) electrons, effectively contribute to the momenta of the centers of mass of the annihilating pairs.

WHILE testing with annihilation radiation a circuit selecting the coincident pulses from two scintillation counters, it was realized that the angular correlation between the two annihilation photons could easily be measured with far greater accuracy than previously reported.<sup>2</sup> As a result, it was considered that a precise measurement of the angular correlation, which would throw some light on the momentum distribution of the centers of mass of the annihilating pairs, and hence, on their mean momentum, was worth attempting. In particular, it was thought of interest to investigate the agreement of the so deduced value for the mean momentum of the centers of mass with the value for the same quantity recently obtained by DuMond, Lind, and Watson from an analysis of the shape of the annihilation  $\gamma$ -ray line.<sup>3</sup> It was also thought of interest to present a theoretical treatment and to compare the results of theory and experiment.

### EXPERIMENTAL ARRANGEMENT AND RESULTS

Two anthracene detectors<sup>4</sup> were used with type 1P-21 photo-multipliers, whose pulses were amplified with amplifiers of  $0.2 \mu\text{sec.}$  rise time, and fed to a coincidence circuit of  $0.3 \mu\text{sec.}$  resolving time.

The pieces of anthracene were in contact with the envelope of the multiplier tubes. In order to minimize the effect of scattering, light tightness was insured by wrapping the tubes with rubber tape, and the pieces of anthracene were covered only with an aluminum foil 1 mil thick. The surfaces of the anthracene samples

located directly under the aluminum foil were plane and their positions could easily be located from the outside.

The source of positrons consisted of thin "shavings" (two or three pieces  $1 \times 3$  mm and a few mils thick) from a deuteron-bombarded copper target, rich in the 14-hour  $\text{Cu}^{64}$  activity. The copper "shavings" were introduced in a small gold tube, obtained by wrapping sixfold a 1 mil gold foil. This tube was subsequently flattened and effectively constituted a source of annihilation gamma-rays of thickness of only a few tenths of a mm, since, as is easily seen, most of the positrons were absorbed in the inner layers of the gold.

Several measurements of the angular distribution were performed with the source at different distances from the detectors. During the final experiments, whose results are shown in Fig. 1, the distance between the source and either detector was 120 cm (Fig. 2). For maximum efficiency and best angular definition, the gamma-rays from the source were made to traverse the detectors along their longest dimension. In these conditions the detectors, seen from the source, subtended an angle of  $4 \times 10^{-3}$  rad. =  $15'$  in the vertical direction, and an angle of  $10^{-2}$  rad. =  $40'$  in the horizontal direction; while the source, seen from the detectors, covered an angle of only a few minutes in the vertical direction and of about  $15'$  in the horizontal direction. The vertical alignment of the apparatus was made by means of a cathetometer and the measurements were performed by displacing one of the detectors vertically. The displacements were measured with the cathetometer from the position where the source and the upper surfaces of the detectors were in the same horizontal line; this position corresponds to the zero on the abscissas of Fig. 1, downward displacement of the detector being taken as positive.

The peak of the curve occurs for a negative displacement of about 0.6 cm, corresponding to the position at which the line joining the centers of the detectors passes through the source. The coincidence rate was  $\approx 60$  c/m at the peak, and  $\approx 1$  c/m at a displacement 3 cm away from the peak. The curve is practically symmetrical about the peak; however, some errors

<sup>1</sup> Preliminary note in Phys. Rev. **76**, 440 (1949).

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<sup>2</sup> R. Beringer and C. G. Montgomery, Phys. Rev. **61**, 222 (1942). A note by I. Pomeranchuk, J. Exp. Theor. Phys. (Russian) **19**, 183 (1949) on the "Mean life of slow positrons," received here after the present work was largely completed, contains a very brief discussion of the theory and mentions experiments by N. A. Vlassov and E. A. Tzirelson (Dokl. Akad. Nauk U.S.S.R. **59**, 879 (1948)) with results apparently in agreement with ours.

<sup>3</sup> DuMond, Lind, and Watson, Phys. Rev. **75**, 1226 (1949).

<sup>4</sup> Kindly supplied by P. R. Bell of the Oak Ridge National Laboratory.

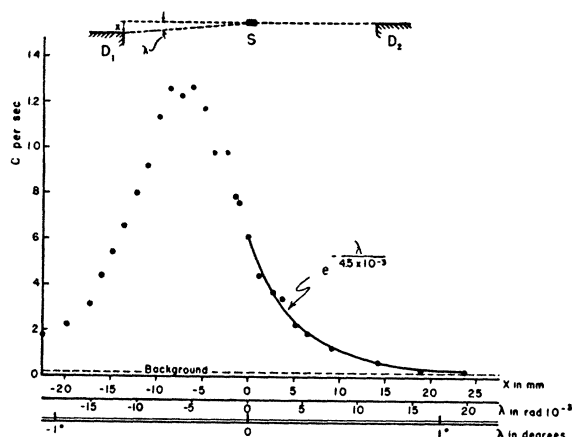


FIG. 1. Photon-photon coincidences as a function of the position of one of the detecting counters.

(due to imperfect alignment of the lower surface of the detectors, to scattering from the multiplier tubes, and to the direct response of the multipliers to gamma-rays) were expected on the negative side, and only the positive (right-hand) side of the peak was carefully studied. The fact that the counting rate does not vanish for positive displacements conclusively indicates that the annihilation gamma-rays are not always emitted in exactly opposite directions.

The possible influence of Compton scattering on the annihilation gamma-rays was studied experimentally by increasing the thickness of the gold absorber and by inserting a number of thin aluminum and paper foils along the path of the gamma-rays. Since the shape of the curve for positive displacements was not sensibly affected, we feel justified in assuming that the effect of Compton scattering can be neglected in the analysis of the observed distribution.<sup>5</sup>

#### THE MEAN MOMENTUM OF THE CENTERS OF MASS OF THE ANNIHILATING PAIRS

Let us call  $\lambda$  the experimentally measured angle between the lines connecting the source with the upper surfaces of the two detectors,  $\omega$  the vertical angular width of the detectors, and  $C(\lambda)$  the observed coincidence counting rate. Let us call  $N(p)dp$  the number of annihilating pairs having center of mass momentum of magnitude between  $p$  and  $p+dp$ . We want to find a relation between the measured quantities and the function  $N(p)$ .

For this purpose let us first consider the function  $N_z(p_z)$  expressing the distribution of the annihilating pairs according to their  $z$  (vertical) components of center of mass momentum, and observe that the horizontal angular aperture of our detectors is, on the one hand, so small that its square can be neglected, but, on the other, still so large compared to the angular aperture of the annihilation quanta that the problem

can be treated in a vertical plane without too large errors. Then, since,  $p_z/mc$  is numerically equal to the vertical angle  $\alpha$  (in radians) measuring the departure from  $180^\circ$  of the annihilation quanta, we can write:

$$C(\lambda) = \text{const.} \int_0^\omega d\beta \int_{\beta+\lambda}^{\beta+\omega+\lambda} N_z(\alpha) d\alpha,$$

so that after double differentiation, and apart from trivial constants,

$$[d^2C(\lambda)]/d\lambda^2 = N_z(2\omega + \lambda) - 2N_z(\omega + \lambda) + N_z(\lambda). \quad (1)$$

After  $N_z(p_z)$  is obtained by means of this relation,  $N(p)$  may be immediately computed from:

$$N(p) = -2p[dN_z(p)/dp].$$

The accuracy of our data hardly justifies the detailed application of this procedure, which will be used only as a guide in what follows. Since the experimentally determined  $C(\lambda)$ , for  $\lambda > 0$ , can be closely approximated by  $(\text{const.}) e^{-\lambda/\lambda_0}$ , with  $\lambda_0 = 4.5 \times 10^{-3}$  rad. (see Fig. 1), solution of the difference Eq. (1) immediately yields

$$\begin{aligned} N_z(\lambda) &= (\text{const.}) \lambda_0^{-2} (e^{-\omega/\lambda_0} - 1)^{-2} e^{-\lambda/\lambda_0} \\ &= (\text{const.}') e^{-\lambda/\lambda_0}. \end{aligned} \quad (2)$$

Thus we can write  $N(p) = (\text{const.}) p \exp(-p/4.5 \times 10^{-3} \text{ mc})$ . We will use this expression only for the determination of the average value of  $p$ , with the result

$$\bar{p}_{\text{Au}} = 2 \times 4.5 \times 10^{-3} \text{ mc} = 1.2 \text{ mc}/137. \quad (3)$$

This value is in order of magnitude agreement with that obtained by DuMond, Lind, and Watson, from an analysis of the shape of the annihilation  $\gamma$ -ray line from a Cu absorber, viz.:

$$\bar{p}_{\text{Cu}} = 8 \times 10^{-3} \text{ mc} = 1.1 \text{ mc}/137. \quad (4)$$

#### THEORETICAL ESTIMATE OF THE MEAN MOMENTUM OF THE CENTERS OF MASS OF THE ANNIHILATING PAIRS

In order to obtain a theoretical estimate of the mean momentum of the centers of mass of the annihilating pairs,  $\bar{p}$ , it is convenient to treat separately the contributions of the electron and of the positron.

Let us consider the positron first. We note that approximately 98 percent of the positrons reach the end of their "path of ionization" without suffering annihilation.<sup>6</sup> At the end of this path the positron kinetic

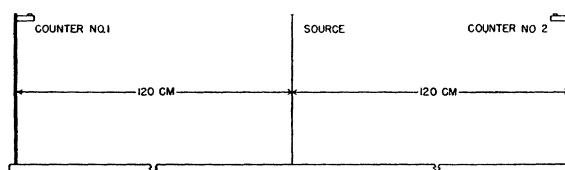


FIG. 2. Location of source and detecting counters in the final experiments.

<sup>5</sup> See also, DuMond, Lind, and Watson, reference 3, and J. W. M. DuMond, Phys. Rev. 75, 1266 (1949).

<sup>6</sup> W. Heitler, *Quantum Theory of Radiation* (Oxford University Press, London, 1944), p. 230 ff.

energy is of the same order of magnitude as that of the fastest electrons in the degenerate electron gas in the metal. Subsequent energy losses for the positron are then possible largely through inelastic collisions with the thermal lattice vibrations of the metal atoms. We shall show in what follows (see Appendix I) that, as a result of such inelastic collisions, the positrons attain thermal equilibrium with the lattice in a time short compared with the mean time for annihilation. Once thermalized, the positrons diffuse randomly through the metal without any further energy gain or loss on the average, and are eventually annihilated.

During their diffusion, the thermalized positrons preferentially occupy interstitial positions in the lattice as a consequence of the Coulomb repulsion of the positive metallic ions. We, therefore, describe the motion of these positrons by a wave-function which is essentially different from zero (and roughly constant), within any atomic polyhedron, only at distances from the nucleus greater than the positive ion radius. Thus, only electrons whose wave functions appreciably extend beyond the ionic volumes will effectively combine with the positrons in the annihilation process.<sup>7</sup> In the present case of Au, such electrons are those occupying the bands arising from the 6s, and, possibly the 5d, atomic orbitals.

We next present arguments (see below) indicating that, to a sufficient approximation for the purpose at hand, we may describe the motion of the 6s electrons by plane wave wave functions outside the ionic volumes, and may neglect the overlap between the 5d electron and the positron wave functions. With the electron and positron wave functions so specified we turn our attention to the matrix element for the two photon annihilation transition; this matrix element is seen, in accordance with radiation theory, to involve the above-mentioned electron and positron wave functions and the plane waves associated with the two emitted photons. The evaluation of the integral for this matrix element then yields, by standard perturbation theory, the probability for the annihilation of an electron and positron of given energies (or rather given wave numbers) with the simultaneous emission of two photons of given total momentum. Finally, we average this annihilation probability over the energy distributions (Fermi and Boltzmann, respectively) of the annihilating electrons and positrons; the result is  $\mathfrak{N}(\mathbf{p})d\mathbf{p}$ , the probability that the annihilation occurs with the total momentum  $\mathbf{p}$  of the two emitted photons lying in the range  $d\mathbf{p}$ . Since  $\mathbf{p}$ , from the conservation of momentum in (any particular) annihilation process, is also the total momentum of the (particular) annihilating electron and positron, i.e., the momentum of the center of mass of the corresponding annihilating pair, a knowledge of  $\mathfrak{N}(\mathbf{p})d\mathbf{p}$  immediately yields the pair's mean center of mass momentum,  $\bar{\mathbf{p}}$ .

<sup>7</sup> See DuMond, Lind, and Watson, reference 3; Pomeranchuk, reference 2.

We may now proceed to obtain quantitative estimates of  $\mathfrak{N}(\mathbf{p})$  and  $\bar{\mathbf{p}}$  in accordance with the scheme outlined in the preceding two paragraphs. First of all, ordinary statistical considerations indicate that:

$$\mathfrak{N}(\mathbf{p})d\mathbf{p} = \text{const.} \sum_{\mathbf{k}_+, \mathbf{k}_-} F_+(\mathbf{k}_+)F_-(\mathbf{k}_-) |\Phi_{\mathbf{k}_+, \mathbf{k}_-}(\mathbf{p})|^2 d\mathbf{p}. \quad (5)$$

In Eq. (5),  $F_{\mp}(\mathbf{k}_{\mp})$  is the probability of the annihilating electron (positron) being in a state with wave number  $\mathbf{k}_{\mp}$ . Thus, since the positrons are supposed thermalized,

$$F_+(\mathbf{k}_+) = (\text{const.}) \exp[-\epsilon_+(\mathbf{k}_+)/\kappa T],$$

while  $F_-(\mathbf{k}_-) = \text{const.}$ , for electron states in the completely filled 1s, 2s,  $\dots$ , 5d bands, and

$$F_-(\mathbf{k}_-) = (\text{const.}) \left\{ \exp\left[\frac{\epsilon_-(\mathbf{k}_-) - \zeta(T)}{\kappa T}\right] + 1 \right\}^{-1}$$

for states in the half-filled 6s band;  $\epsilon_{\mp}(\mathbf{k}_{\mp}) \approx \hbar^2 k_{\mp}^2 / 2m$  is the energy of the  $\mathbf{k}_{\mp}$  states (reckoned relative to the corresponding bottom-of-the-band zero-point energy).<sup>8</sup> The quantity  $\Phi_{\mathbf{k}_+, \mathbf{k}_-}(\mathbf{p})$  is, apart from a constant, the matrix element (i.e. essentially the probability amplitude) for the emission of two photons with total momentum  $\mathbf{p}$ , as a result of the annihilation of an electron and a positron with wave numbers  $\mathbf{k}_-$ ,  $\mathbf{k}_+$ .

Further, radiation theory indicates that (see Appendix II)

$$\Phi_{\mathbf{k}_+, \mathbf{k}_-}(\mathbf{p}) = \int \psi_{\mathbf{k}_+}^*(\mathbf{r}) \psi_{\mathbf{k}_-}(\mathbf{r}) \exp(-i\mathbf{p} \cdot \mathbf{r}) d\mathbf{r}. \quad (6)^9$$

or, alternatively,

$$\Phi_{\mathbf{k}_+, \mathbf{k}_-}(\mathbf{p}) = (2\pi)^3 \int \varphi_{\mathbf{k}_+}^*(\mathbf{p}_+) \varphi_{\mathbf{k}_-}(\mathbf{p}_-) \times \delta(\mathbf{p}_+ + \mathbf{p}_- - \mathbf{p}) d\mathbf{p}_+ d\mathbf{p}_-. \quad (7)$$

In Eqs. (6), (7),  $\psi_{\mathbf{k}_{\mp}}(\mathbf{r})$ ,  $\varphi_{\mathbf{k}_{\mp}}(\mathbf{r})$  are, respectively, the coordinate space and the momentum space wave functions of the annihilating electron and positron, appropriate to states of motion in the periodic potential of the lattice specified by wave number  $\mathbf{k}_{\mp}$ . The  $\delta$  function in the integral of Eq. (7) expresses the conservation of momentum in any particular annihilation and justifies the additional interpretation of  $\Phi_{\mathbf{k}_+, \mathbf{k}_-}(\mathbf{p})$  as the probability amplitude for the center of mass of the annihilating electron and positron (with wave numbers,  $\mathbf{k}_-$ ,  $\mathbf{k}_+$ ) having the momentum value  $\mathbf{p}$ .

<sup>8</sup> To a sufficient approximation, we may take throughout, as the effective crystal lattice mass of both the valence 6s electron and the positron, the free electron mass,  $m$ . This assumption is roughly justified, for the electron by the work of Krutter in reference 13 on the homologous case of Cu, and for the positron, by Eq. (23)-(25), below. It may also be mentioned that the general description used of the motion of a charged particle in the crystal lattice, the Bloch model, is an even better approximation for the positron than for the valence electron. This follows because of the absence, in the positron case, of exchange interactions (in the ordinary sense) with the electrons of the solid.

<sup>9</sup> Wave number units are used for the momenta  $\mathbf{p}$ ,  $\mathbf{p}_-$ ,  $\mathbf{p}_+$ .

To evaluate the integrals in Eq. (6) or in Eq. (7) for  $\Phi_{\mathbf{k}_+, \mathbf{k}_-}(\mathbf{p})$  one recalls the Fourier expansions of coordinate space and momentum space wave functions in a periodic potential:

$$\psi_{\mathbf{k}\mp}(\mathbf{r}) = \exp(i\mathbf{k}\mp \cdot \mathbf{r}) \sum_{\mathbf{g}\mp} v^{-1} \left[ \int_{\mathbf{r}'} \psi_{\mathbf{k}\mp}(\mathbf{r}') \times \exp[-i(\mathbf{k}\mp \cdot \mathbf{r}' + 2\pi\mathbf{g}\mp \cdot \mathbf{r}')] d\mathbf{r}' \right] \times \exp(2\pi i\mathbf{g}\mp \cdot \mathbf{r}), \quad (8)$$

$$\varphi_{\mathbf{k}\mp}(\mathbf{p}\mp) = \sum_{\mathbf{g}\mp} v^{-1} \left[ \int_{\mathbf{r}'} \psi_{\mathbf{k}\mp}(\mathbf{r}') \times \exp[-i(\mathbf{k}\mp \cdot \mathbf{r}' + 2\pi\mathbf{g}\mp \cdot \mathbf{r}')] d\mathbf{r}' \right] \times \delta(\mathbf{k}\mp + 2\pi\mathbf{g}\mp - \mathbf{p}\mp), \quad (9)$$

$v$ , and  $\mathbf{g}_-$ ,  $\mathbf{g}_+$ ,  $\mathbf{g}$ , being the volume of a unit cell, and reciprocal lattice vectors, respectively. Substituting Eq. (9) into Eq. (7), one then obtains:

$$\Phi_{\mathbf{k}_+, \mathbf{k}_-}(\mathbf{p}) = (2\pi)^3 \sum_{\mathbf{g}} v^{-1} \left[ \int_{\mathbf{r}'} \psi_{\mathbf{k}_+}^*(\mathbf{r}') \psi_{\mathbf{k}_-}(\mathbf{r}') \times \exp[-i(\mathbf{k}_+ + \mathbf{k}_- + 2\pi\mathbf{g} \cdot \mathbf{r}') d\mathbf{r}'] \right] \times \delta(\mathbf{k}_+ + \mathbf{k}_- + 2\pi\mathbf{g} - \mathbf{p}). \quad (10)$$

As already noted above, the integral in Eq. (10) for  $\Phi_{\mathbf{k}_+, \mathbf{k}_-}(\mathbf{p})$  indicates that the annihilation probability effectively vanishes for all electrons except those in the 6s and, perhaps, the 5d bands, since it is only for these last that the corresponding  $\psi_{\mathbf{k}_-}(\mathbf{r})$  can overlap appreciably with the  $\psi_{\mathbf{k}_+}(\mathbf{r})$ .

We must now specify quantitatively the 6s and the 5d electron wave functions and the positron wave function within a unit cell of the metal's crystal lattice. In accord with our previous assumption about the positron wave function and with the general method of Wigner and Seitz,<sup>10</sup> we may take:

$$\psi_{\mathbf{k}_+}(\mathbf{r}) \exp(-i\mathbf{k}_+ \cdot \mathbf{r}) \quad (a)$$

spherically symmetric within the atomic polyhedron constituting the unit cell;

$$\psi_{\mathbf{k}_+}(\mathbf{r}) \exp(-i\mathbf{k}_+ \cdot \mathbf{r}) = 0 \text{ for } r \leq r_i; \quad (b)$$

$$\partial/\partial r [\psi_{\mathbf{k}_+}(\mathbf{r}) \exp(-i\mathbf{k}_+ \cdot \mathbf{r})] = 0 \text{ at } r = r_p; \quad (c)$$

$r_i$  and  $r_p$  are, respectively, the radii of the Au<sup>+</sup> ion and of a sphere equivoluminal with the atomic polyhedron. The conditions (a), (b), (c), yield for

<sup>10</sup> E. Wigner and F. Seitz, Phys. Rev. 43, 804 (1933); 46, 509 (1934).

$$r_i < r < r_p,$$

$$\psi_{\mathbf{k}_+}(\mathbf{r}) \exp(-i\mathbf{k}_+ \cdot \mathbf{r}) = (\text{const.}) \sin[\alpha(r - r_i)]/\alpha r$$

with  $\alpha r_p = \tan[\alpha(r_p - r_i)]$ ,<sup>11</sup> which, for  $(r_i)_{\text{Au}} \approx 1.0 \text{ \AA}$  (see footnote 17), and  $(r_p)_{\text{Au}} \approx 1.6 \text{ \AA}$ , is not too badly approximated by replacing  $\sin[\alpha(r - r_i)]/\alpha r$  by 1. This last approximation, which is adopted below, may actually be closer to the true  $\psi_{\mathbf{k}_+}(\mathbf{r})$  than the previous expression since: (a) the outer shells of the ion cannot be considered as absolutely impenetrable to the positron, and (b) once the positron does penetrate the ion more deeply it encounters an effective nuclear repulsion increasing much faster than (distance)<sup>-2</sup>; both (a) and (b) tend to produce a more precipitate variation of  $\psi_{\mathbf{k}_+}(\mathbf{r}) \exp(-i\mathbf{k}_+ \cdot \mathbf{r})$  for  $r \approx r_i$  than is described by  $\sin[\alpha(r - r_i)]/\alpha r$ ,

As regards the Au 6s electron wave function, Wigner-Seitz-Slater type<sup>12</sup> calculations by Krutter<sup>13</sup> and by Tibbs<sup>14</sup> for the homologous case of Cu, indicate that for  $r_i < r < r_p$ , i.e. outside the Au<sup>+</sup> ion,  $\psi_{\mathbf{k}_-}(\mathbf{r})$  will be quite well approximated by the free electron solution:  $(\text{const.}) \exp(-i\mathbf{k}_- \cdot \mathbf{r})$ . Within the ion ( $r < r_i$ ) the form of  $\psi_{\mathbf{k}_-}(\mathbf{r})$  approximates to an isolated atom 6s (+5d + ...) wave function, but in any case, is irrelevant for our calculation; this follows from the effective vanishing of  $\psi_{\mathbf{k}_+}(\mathbf{r})$  for  $r < r_i$  and the resultant absence of electron-positron wave function overlap in the ion interior, in the integral of Eq. (10) for  $\Phi_{\mathbf{k}_+, \mathbf{k}_-}(\mathbf{p})$ .

It remains to determine the Au 5d electron wave functions. Krutter's calculations for Cu<sup>13</sup> indicate that the energy vs. wave number dependence for the electrons in the 3d band is rather more appropriate to tightly than to loosely bound electrons and so imply a form for  $\psi_{\mathbf{k}_-}(\mathbf{r})$  (within any particular atomic polyhedron) not too different from that for an isolated atom 3d wave function.<sup>14a</sup> Now in an isolated Cu<sup>+</sup> ion, Hartree type calculations show rather small extension of the 3d wave function beyond the ion radius;<sup>15</sup> in the absence of definite information to the contrary we assume that the same situation holds in the analogous case of the 5d electrons in an isolated Au<sup>+</sup> ion and, so, in an Au<sup>+</sup> ion in the metal. It would then seem not too poor an approximation to suppose a very small effective overlap between  $[\psi_{\mathbf{k}_-}(\mathbf{r})]_{5d}$  and  $\psi_{\mathbf{k}_+}(\mathbf{r})$ , and hence to neglect completely the 5d electron contribution to the annihilation probability. This procedure should at least give a reasonable lower limit to the theoretical  $\bar{p}_{\text{Au}}$  and really cannot be avoided in the absence of any actual calculations of the Au metallic 5d wave functions.

<sup>11</sup> In accordance with the general theory of reference 10 one has: positron energy in state  $\psi_{\mathbf{k}_+} \approx (\hbar^2\alpha^2/2m) + (\hbar^2k_+^2/2m)$ .

<sup>12</sup> See reference 10 and J. C. Slater, Phys. Rev. 45, 794 (1934); Rev. Mod. Phys. 6, 209 (1934).

<sup>13</sup> H. M. Krutter, Phys. Rev. 48, 664 (1935).

<sup>14</sup> S. R. Tibbs, Proc. Camb. Phil. Soc. 34, 89 (1938).

<sup>14a</sup> Note added in proof.—A similar implication may be drawn from an examination of Fig. 1 in a note of Steinberger and Wick (Phys. Rev. 76, 994 (1949)) where plots are given of the 3d electron wave function in an isolated Fe atom and in metallic Fe.

<sup>15</sup> D. R. Hartree, Proc. Roy. Soc. 141, 282 (1933); see particularly Table III on p. 297.

Our  $\psi_{\mathbf{k}_+}$  and  $\psi_{\mathbf{k}_-}$  having been specified, it is now a straightforward matter to evaluate the integral in Eq. (10) for  $\Phi_{\mathbf{k}_+, \mathbf{k}_-}(\mathbf{p})$ , and, then, the sums in Eq. (5) for  $\mathfrak{N}(\mathbf{p})$ . We obtain:

$$\Phi_{\mathbf{k}_+, \mathbf{k}_-}(\mathbf{p}) = \text{const.} \sum_{\mathbf{g}} \left[ \delta_{\mathbf{g}, 0} - \frac{v_i}{v} \left\{ \left[ \sin(2\pi g r_i) - (2\pi g r_i) \cos(2\pi g r_i) \right] \frac{3}{(2\pi g r_i)^3} \right\} \right] \times \delta(\mathbf{k}_+ + \mathbf{k}_- + 2\pi\mathbf{g} - \mathbf{p}) \quad (11)$$

and

$$N(\mathbf{p}) = 4\pi p^2 \mathfrak{N}(\mathbf{p}) \approx \text{const.} 4\pi p^2 \left[ \eta_{k_{\max}}(p) \left( 1 - \frac{2v_i}{v} \right) + \frac{1}{2} \left( \frac{v_i}{v} \right)^2 \exp\left( -\frac{2}{\pi^2} r_i^2 p^2 \right) \right], \quad (12)$$

several purely mathematical approximations being introduced to carry out the sums over  $\mathbf{k}_-$  and  $\mathbf{k}_+$  in Eq. (5).<sup>16</sup> In Eqs. (11), (12),  $v_i$  and  $v$  are, respectively, the volumes of the  $\text{Au}^+$  ion and the Au atomic polyhedron (unit cell);  $k_{\max} = 2\pi(3/(8\pi v))^{1/3}$  is the maximum value of  $|\mathbf{k}_-|$  for an electron in the half-filled 6s band (at  $T=0$ );

$$\eta_{k_{\max}}(p) = 1 \text{ if } p < k_{\max} \text{ and } \eta_{k_{\max}}(p) = 0 \text{ if } p > k_{\max}.$$

As in the case of the experimental  $N(p)$  distribution, we shall use the (qualitatively similar) theoretical  $N(p)$  distribution only to calculate the mean momentum of the annihilating pair centers of mass. Equation (12) then yields,

$$\bar{p} = \int_0^\infty p N(p) dp / \int_0^\infty N(p) dp = \frac{3}{4} - \hbar k_{\max} \left[ \frac{1 - 2v_i/v + 1.80(v_i/v)^{1/2}}{1 - 2v_i/v + 1.03(v_i/v)} \right], \quad (13)$$

whence, using  $v=1/4$  (Au lattice constant)<sup>3</sup> =  $1/4$  ( $4.07 \text{ \AA}^\circ$ )<sup>3</sup> and  $v_i/v=1/4$  ( $r_i=1.0 \text{ \AA}^\circ$ ), we obtain:<sup>17</sup>

$$\bar{p}_{\text{Au}} = \frac{3}{4} \hbar k_{\max} [1.6] = 0.8 \text{ mc}/137. \quad (14)$$

<sup>16</sup> These mathematical approximations break down if  $v_i/v$  is too close to  $\frac{1}{2}$  (or between  $\frac{1}{2}$  and 1). Another method must then be used. It should also be noted that strictly speaking, the  $N(p)$  of Eq. (12) is the annihilating pair center of mass momentum distribution calculated at the absolute zero of temperature; i.e. we have taken  $F_+(\mathbf{k}_+) = (\text{const})\delta(\mathbf{k}_+)$  and  $F_-(\mathbf{k}_-) = (\text{const})\eta_{k_{\max}}(\mathbf{k}_-)$ . The  $N(p)$  distribution at room temperature however is not very different from that at  $T=0$  since  $\kappa T_{\text{room}} \ll \hbar^2 k_{\max}^2 / 2m$ .

<sup>17</sup> The numerical value of  $r_i$  which we employ is obtained from the location of the 5d electron charge density maximum in  $\text{Au}^+$  by using Slater's (isolated atom) hydrogenic wave functions. See J. C. Slater, Phys. Rev. **36**, 57 (1930). However, as emphasized by H. Jones (Physica **15**, 13 (1949)) the large excess of the observed value of the bulk modulus in Au over that calculated from

It will be noted from Eqs. (12), (13), that  $\frac{3}{4}\hbar k_{\max}$  (0.5 mc/137 for Au) is the value of  $\bar{p}_{\text{Au}}$  obtained in the limit  $v_i/v \rightarrow 0$  i.e., obtained on the assumption that both the 6s electron and the positron have wave functions which are of the free particle form:

$$\psi_{\mathbf{k}\mp}(\mathbf{r}) = (\text{const.}) \exp(i\mathbf{k}\mp \cdot \mathbf{r})$$

throughout the whole volume of the metal (including the ionic interiors). Thus, the factor in the square brackets in Eqs. (13), (14), arises from the deviation of  $\psi_{\mathbf{k}\mp}(\mathbf{r})$  from the free particle form, i.e., from the approach of  $\psi_{\mathbf{k}_-}(\mathbf{r})$  to the isolated atom 6s wave function within the ionic interior and from the effective vanishing of  $\psi_{\mathbf{k}_+}(\mathbf{r})$  in this interior. In accordance with the comment made in footnote 16, practically the whole observed value of  $\bar{p}$  at room temperature arises from the zero-point motion of the electrons and the positrons, the temperature variation of  $\bar{p}$  being very slight

$$\langle \langle p(T) \rangle_{\text{Av}} \rangle = \langle p(0) \rangle_{\text{Av}} [1 + \gamma(\kappa T / \hbar^2 k_{\max}^2 / 2m)^2] \text{ with } \gamma \sim 1.$$

A point worthy of examination is the possible effect on the annihilation photon angular distribution of the so far neglected Coulomb attraction between the members of the annihilating pair. It would seem that there is no such effect, at least in first order, since the operator corresponding to the pair's center of mass momentum commutes with the pair's Coulomb energy operator.<sup>18</sup> For the sake of completeness, we should also mention the possibility of the formation of positronium atoms within the solid with an ultimate two photon self-annihilation out of their ground  $^1S$  states.<sup>19</sup> Here it appears possible that the angular distribution of the two annihilation photons might be shifted in a direction appropriate to smaller apparent electron-positron center of mass momenta. Such a shift will occur if, (a): the center of mass momentum of the positronium atom upon formation, (equal to the vector sum of the momenta of the combining electron and positron

the  $\epsilon_-(\mathbf{k}_-)$  vs.  $\mathbf{k}_-$  dependence for the 6s valence electrons, indicates a relatively large (repulsive) interionic overlap energy and hence a relatively large value of  $v_i/v$ , probably  $> \frac{1}{4}$ . Such a value of  $v_i/v$  would increase the theoretical value of  $\bar{p}_{\text{Au}}$  (Eqs. (13), (14)) and would thus improve its agreement with the experimental value (Eq. (3)).

<sup>18</sup> This Coulomb attraction may, however, be expected to decrease somewhat the positron mean life for annihilation by increasing the electron density near the positron. See Pomeranchuk, reference 2; J. W. Shearer and M. Deutsch, Phys. Rev. **76**, 462 (1949), and the comment in footnote 31.

<sup>19</sup> J. Pirenne, Arch. D. Sci. Phys. et Natur. **28**, 273 (1946); **29**, 121 (1947); J. A. Wheeler, Ann. New York Acad. Sci. **48**, 219 (1946); A. Ore and J. L. Powell, Phys. Rev. **75**, 1696 (1949); L. D. Landau, Dokl. Akad. Nauk. U.S.S.R. **60**, 207 (1948); E. M. Lifshitz, Dokl. Akad. Nauk. U.S.S.R. **60**, 211 (1948); I. Pomeranchuk, Dokl. Akad. Nauk. U.S.S.R. **60**, 213 (1948). These authors also show that if any positronium annihilates itself out of the ground  $^3S$  state, three photons are simultaneously emitted. Further Ore and Powell show that the ratio of the total cross sections for three and for two photon annihilation of slowly moving (but mutually unbound) electrons and positrons is  $\approx 1/370$ . Remembering in addition, the roughly isotropic three photon angular distribution and the solid angle factor, it is seen that the contribution of the three photon process to the observed breadth (and background) of the two photon angular distribution ( $C(\lambda)$  of Eq. (11)) must be completely negligible.

$\sim mc/137$ ), is decreased to values  $\sim(4m\kappa T)^{\frac{1}{2}}$  by thermalizing collisions with lattice atom vibrations, in a time short compared with that required for annihilation,<sup>20</sup> and (b), the positronium atom is not dissociated in any of these collisions. Item (b), however, is rather doubtful since the positronium atom in the crystal lattice probably has a very small positive or even a negative binding energy and is thus quite unstable. This last conclusion follows from the fact that the (isolated) positronium first Bohr orbit diameter ( $2 A^\circ$ ) is not much smaller than the distance between nearest neighbor Au lattice ions; the positronium wave function is then greatly compressed to enable the atom to fit into and move through the lattice, and such a compression involves a very large decrease in binding energy.<sup>21</sup>

### DISCUSSION

The agreement between the experimental value and the (lower limit on the) theoretical value of  $\bar{p}_{Au}$  (Eqs. (3) and (14)) is perhaps as good as can be expected considering the rough and preliminary nature of both. Several obvious uncertainties affect our theoretical formulas, in particular a somewhat dubious numerical value for  $v_i/v$  ( $=1/4$ ) has been used (see comment in footnote 17). On the other hand, our expression for  $\bar{p}$  (Eq. (13)) is not too sensitively dependent on the necessarily somewhat arbitrary magnitude of  $v_i/v$ ; values of this ratio of 1/5 and 1/3, for example, yield values of  $\bar{p}_{Au}$  of 0.75  $mc/137$  and 0.9  $mc/137$ , respectively. Nevertheless it is quite clear that any really quantitatively reliable values of  $\bar{p}$  and  $N(\bar{p})$  will necessitate Wigner-Seitz type calculations (with the proper ion core fields in the individual atomic polyhedra) for the appropriate electron and positron wave functions.

Certain additional experiments are immediately suggested by the general form of Eq. (13) for  $\bar{p}$ . Thus, if one should use as positron absorbers the members of a chemically homologous series of solids possessing the same crystal structure, e.g., the various alkali metals; Li, Na, K, Rb, Cs, one could expect about the same value of  $v_i/v$  for the different members of the series,<sup>21a</sup> so that their  $\bar{p}$  would vary directly with their  $k_{max}$  or inversely with their lattice constant. Since the lattice constant increases by a factor of 1.75 from Li to Cs, the corresponding decrease of  $\bar{p}$  should be easily observable. Another interesting test would involve the use of two series of alkali halides, e.g.: (a)  $Li^+Cl^-$ ,  $Na^+Cl^-$ ,  $K^+Cl^-$ ,  $Rb^+Cl^-$ , and (b)  $Na^+F^-$ ,  $Na^+Cl^-$ ,  $Na^+Br^-$ ,

<sup>20</sup> The ground  $1S$  state annihilation of positronium has a mean life of  $1.25 \times 10^{-10}$  sec. (see reference 19).

<sup>21</sup> Thus, A. Sommerfeld and H. Welker, *Ann. d. Physik* **32**, 56 (1938), show that the lowest discrete energy of the H-atom becomes positive if the atom is confined in an impenetrable sphere centered at the atom's center of mass and having a radius  $\leq 1.835$ , the radius of the (isolated) H-atom Bohr orbit.

<sup>21a</sup> For a discussion and criticism of such an expectation, see e.g. F. Seitz, *Modern Theory of Solids* (McGraw-Hill Book Company, Inc., New York, 1940), Chapter 2.

$Na^+I^-$ . Then, since one expects that, to a first approximation, the positron annihilates only against the fairly tightly bound outer shell electrons of the negatively charged halogen ions, and has a wave function immediately before annihilation determined mainly by the electrostatic potential of these ions, one should find about the same value of  $\bar{p}$  for all the members of the (a) series and a monotonic variation in  $\bar{p}$  along the (b) series. In general, it is clear that the study of the angular distribution of annihilation radiation with scintillation counters in coincidence will help throw light on electronic momentum distributions, in the atoms and molecules of gases and in liquids and solids.

### APPENDIX I

#### Thermalization Time of the Positron

In conclusion, we shall present an estimate of the time interval  $\tau$ , required for the thermalization of positrons in a metallic solid. It will be recalled that practically all the positrons survive the atom ionization (and excitation) process without annihilation; then, having ceased to lose energy by kicking electrons from the half-filled  $6s$  and the all-filled  $5d$ ,  $5p$ ,  $\dots$ ,  $2s$ ,  $1s$  bands into unoccupied states in the overlapping  $6s$ ,  $6p$ ,  $\dots$  bands ( $p_+ < \sim mc/137$ ), the positrons begin to lose energy by collisions with the thermally vibrating lattice atoms (or ions) exciting these into states of higher vibrational energy. The net rate of positron energy loss as a result of such collisions is given, on the average, by

$$-\frac{d}{dt}\epsilon_+(\mathbf{k}_+) = \sum_{\mathbf{g}} \int \frac{d\boldsymbol{\sigma}}{(2\pi)^3} \left\{ \frac{2\pi}{\hbar} \left| \langle \mathbf{k}_+ | H_{inter} | \mathbf{k}_+ - \boldsymbol{\sigma} - 2\pi\mathbf{g} \rangle \right|^2 \right. \\ \times \delta(\epsilon_+(\mathbf{k}_+) - \epsilon_+(\mathbf{k}_+ - \boldsymbol{\sigma} - 2\pi\mathbf{g}) - \hbar\omega(\boldsymbol{\sigma})) \\ \left. - \frac{2\pi}{\hbar} \left| \langle \mathbf{k}_+ | H_{inter} | \mathbf{k}_+ + \boldsymbol{\sigma} + 2\pi\mathbf{g} \rangle \right|^2 \right. \\ \left. \times \delta(\epsilon_+(\mathbf{k}_+) - \epsilon_+(\mathbf{k}_+ + \boldsymbol{\sigma} + 2\pi\mathbf{g}) + \hbar\omega(\boldsymbol{\sigma})) \right\} \hbar\omega(\boldsymbol{\sigma}). \quad (15)$$

In Eq. (15), the first term describes the mean rate of positron energy loss in collisions in which a vibrational quantum of wave number  $\boldsymbol{\sigma}$  and energy  $\hbar\omega(\boldsymbol{\sigma})$  is transferred from the positron to the lattice vibration, the positron making a transition from a state  $\psi_{\mathbf{k}_+}$  to a state  $\psi_{\mathbf{k}_+'} = \psi_{\mathbf{k}_+ - \boldsymbol{\sigma} - 2\pi\mathbf{g}}$ ;  $\mathbf{g}$ , as before, is a reciprocal lattice vector, the transitions to the states  $\psi_{\mathbf{k}_+'}$  mentioned being the only allowed by the linear momentum conservation selection rule.<sup>22</sup> In a similar way, the second term in Eq. (15) describes the mean rate of positron energy gain in the "inverse" collisions where a vibrational quantum is transferred from the lattice vibration to the positron. The transition probabilities for both these types of collisions are given by the absolute squares of the corresponding matrix elements,

$$|\langle \mathbf{k}_+ | H_{inter} | \mathbf{k}_+ \mp \boldsymbol{\sigma} \mp 2\pi\mathbf{g} \rangle|^2,$$

multiplied by  $2\pi/\hbar$  and by the appropriate  $\delta$  functions which maintain the conservation of (unperturbed) energy in the collisions.

The matrix elements in Eq. (15) may be expressed in accordance with the usual methods of the theory of metals,<sup>23</sup> by

$$\langle \mathbf{k}_+ | H_{inter} | \mathbf{k}_+ \mp \boldsymbol{\sigma} \mp 2\pi\mathbf{g} \rangle = \left[ \int \psi_{\mathbf{k}_+'}^*(\mathbf{r}) \{ \exp(\pm i\boldsymbol{\sigma} \cdot \mathbf{r}) \mathbf{a}_{\boldsymbol{\sigma}} \cdot \text{grad} V(\mathbf{r}) \} \right. \\ \left. \times \psi_{\mathbf{k}_+ \mp \boldsymbol{\sigma} \mp 2\pi\mathbf{g}}(\mathbf{r}) d\mathbf{r} \right] v^{\frac{1}{2}} \left( \frac{\hbar}{2M\omega(\boldsymbol{\sigma})} \left[ \frac{n(\boldsymbol{\sigma})+1}{\text{or } n(\boldsymbol{\sigma})} \right] \right)^{\frac{1}{2}} \quad (16)$$

<sup>22</sup> See Eq. (9), where  $\varphi_{\mathbf{k}_+}(\mathbf{p}_+)$  and  $\varphi_{\mathbf{k}_+'}(\mathbf{p}_+')$  vanish unless  $\mathbf{p}_+ = \mathbf{p}_+ + 2\pi\mathbf{g}_+$ ,  $\mathbf{p}_+' = \mathbf{k}_+' + 2\pi\mathbf{g}_+'.$  Then, since one must have  $\mathbf{p}_+' = \mathbf{p}_+ \mp \boldsymbol{\sigma}$ , one immediately obtains  $\mathbf{k}_+' = \mathbf{k}_+ \mp \boldsymbol{\sigma} + 2\pi(\mathbf{g}_+ - \mathbf{g}_+')$ .

<sup>23</sup> F. Bloch, *Zeits. f. Physik* **52**, 555 (1928); A. Sommerfeld and H. A. Bethe, *Handbuch der Physik* **24**, II (J. Springer, Berlin,

the positron—lattice interaction potential,  $H_{\text{inter}}$ , being taken appropriate to the Bloch deformable ion approximation<sup>24</sup> as

$$v^{\dagger} \mathbf{A}(\boldsymbol{\sigma}) \exp(i\boldsymbol{\sigma} \cdot \mathbf{r}) \text{grad} V(\mathbf{r}) + \text{comp. conj.}$$

Here,  $V(\mathbf{r})$  is the time average (static) periodic potential of the lattice.  $\mathbf{A}(\boldsymbol{\sigma})$  is the  $\boldsymbol{\sigma}$ -normal mode lattice vibration amplitude. The quantity  $v^{\dagger}$  appears because of our present normalization:  $\int_{\sigma} |\psi_{\mathbf{k}+\boldsymbol{\sigma}}(\mathbf{r})|^2 d\mathbf{r} = 1$ . The square root in Eq. (16) times  $\mathbf{a}_{\boldsymbol{\sigma}}$ , the  $\boldsymbol{\sigma}$ -normal mode polarization unit vector, is just the matrix element of  $\mathbf{A}(\boldsymbol{\sigma})$  with respect to the vibrational eigenfunctions in transitions in which a vibration quantum is created or destroyed; in this square root,  $M$  is the vibrating atom mass and  $n(\boldsymbol{\sigma}) = [\exp(\hbar\omega(\boldsymbol{\sigma})/\kappa T) - 1]^{-1}$  is the number of vibrational quanta of energy  $\hbar\omega(\boldsymbol{\sigma})$  and wave number  $\boldsymbol{\sigma}$  present just before the occurrence of the transition.

In the case  $\mathbf{g} = 0$ , the integrals in the matrix elements of Eq. (16) may be transformed by an integration by parts and use of the Schrödinger equation satisfied by  $\psi_{\mathbf{k}+\boldsymbol{\sigma}}(\mathbf{r})$ ; assuming in addition that in a given atomic polyhedron (unit cell)  $\psi_{\mathbf{k}+\boldsymbol{\sigma}}(\mathbf{r}) \exp(-i\mathbf{k}_+ \cdot \mathbf{r})$  satisfies the Wigner-Seitz boundary conditions, is spherically symmetric, and depends only weakly on  $\mathbf{k}_+$ , one obtains,<sup>25</sup>

$$\begin{aligned} & \int_{\sigma} \psi_{\mathbf{k}+\boldsymbol{\sigma}}^*(\mathbf{r}) [\exp(\pm i\boldsymbol{\sigma} \cdot \mathbf{r}) \mathbf{a}_{\boldsymbol{\sigma}} \cdot \text{grad} V(\mathbf{r})] \psi_{\mathbf{k} \mp \boldsymbol{\sigma}}(\mathbf{r}) d\mathbf{r} \\ &= \pm \frac{2}{3} i\sigma \left[ \frac{\hbar^2}{2m} \int_{\sigma} |\text{grad} \psi_{\mathbf{k}+\boldsymbol{\sigma}}(\mathbf{r}) \exp(-i\mathbf{k}_+ \cdot \mathbf{r})|^2 d\mathbf{r} \right] \\ &= \frac{2i}{3} \sigma \left( \frac{\langle p_+^2 \rangle_{\text{Av}}}{2m} \right), \end{aligned} \quad (17)$$

where  $\langle p_+^2 \rangle_{\text{Av}}$  is the mean square positron momentum in the state with  $\mathbf{k}_+ = 0$ , so that  $\langle p_+^2 \rangle_{\text{Av}}/2m$  is just the positron's bottom-of-the-band zero-point kinetic energy. On the other hand, when  $\mathbf{g} \neq 0$ , it does not seem possible to obtain a simplified expression for the matrix elements in Eq. (16). We confine ourselves, therefore, to the remark that in this case, the oscillating factor  $\exp(\mp 2\pi i \mathbf{g} \cdot \mathbf{r})$  in the integral over  $v$  (arising from  $\psi_{\mathbf{k}+\boldsymbol{\sigma}} \mp \boldsymbol{\sigma} \mp 2\pi \mathbf{g}$ ) will tend to decrease this integral's value (at least, for fairly large  $\mathbf{g}$ ); in addition, the energy and momentum conservation laws governing the collisions, yield:<sup>26</sup>

$$1 \geq \left| \frac{\mathbf{k}_+ \cdot (\boldsymbol{\sigma} + 2\pi \mathbf{g})}{|\mathbf{k}_+| |\boldsymbol{\sigma} + 2\pi \mathbf{g}|} \right| = \left| \frac{|\boldsymbol{\sigma} + 2\pi \mathbf{g}|}{2|\mathbf{k}_+|} \pm \frac{m\omega(\boldsymbol{\sigma})}{\hbar |\mathbf{k}_+| |\boldsymbol{\sigma} + 2\pi \mathbf{g}|} \right|$$

and this condition cannot be satisfied, even for the smallest non-vanishing  $2\pi \mathbf{g}$ , once  $\hbar k_+$  becomes smaller than  $\approx \frac{1}{2} \hbar \sigma_{\text{max}}$ <sup>27</sup> (which is,  $\approx \frac{1}{2} mc/137$  for Au). Thus it would seem as if our subsequent complete neglect of the  $\mathbf{g} \neq 0$  collisions will yield a positron (net) average energy loss correct for  $(2m\kappa T)^{\frac{1}{2}} < \hbar k_+ < \approx \frac{1}{2} mc/137$  and somewhat underestimated for  $\hbar k_+ > \frac{1}{2} mc/137$ ; since most of the time required for thermalization, however, is spent in the range of positron momenta from  $\frac{1}{2} mc/137$  to  $(2m\kappa T)^{\frac{1}{2}}$  (see footnote 30) it is clear that the ultimate overestimate of  $\tau$  will not be large.

1933); J. Bardeen, Phys. Rev. **52**, 688 (1937); F. Seitz, Phys. Rev. **73**, 549 (1948). In Seitz's paper, Peierls' criterion for the validity of the general type of time-dependent perturbation theory used, is given in convenient form. It may be verified that this criterion is satisfied in the situation which we consider.

<sup>24</sup> See particularly J. Bardeen, reference 23.

<sup>25</sup> See A. Sommerfeld and H. A. Bethe, reference 23, pp. 512, 513, for the wholly analogous calculation in the case of metallic electronic conduction.

<sup>26</sup> This condition gives those  $\boldsymbol{\sigma}$  and  $\mathbf{g}$  values for which the argument of the  $\delta$  functions in Eq. (15) vanishes, and which, therefore, specify the allowed collisions that make a contribution to the  $\Sigma_{\mathbf{g}} \int d\boldsymbol{\sigma} \dots$ .

<sup>27</sup> This conclusion is a consequence of the fact that, for example, in a face centered cubic lattice like Au,

$$2\pi g_{\text{min}} = \frac{2\pi\sqrt{3}}{(4v)^{\frac{1}{2}}} > \sigma_{\text{max}} = \frac{2\pi}{(4v)^{\frac{1}{2}}} \left( \frac{3}{\pi} \right)^{\frac{1}{2}}$$

and so, the first term on the right-hand side of the inequality will necessarily be greater than 1, for all possible  $\boldsymbol{\sigma}$  and  $2\pi \mathbf{g} \neq 0$  if  $k_+ < \frac{1}{2}(\sqrt{3} - (3/\pi)^{\frac{1}{2}})\sigma_{\text{max}}$ . As regards the second term, it is numerically negligible for all  $\hbar k_+$  of interest [ $\hbar k_+ \geq (2m\kappa T)^{\frac{1}{2}}$ ].

We may now use Eqs. (17) and (16) for the collision matrix elements in the energy loss Eq. (15). With  $\cos\theta_{\sigma} \equiv \mathbf{k} \cdot \boldsymbol{\sigma}/k\sigma$ , one has,<sup>28</sup>

$$\begin{aligned} -\frac{d}{dt} \epsilon_+(\mathbf{k}_+) &= \int_0^{\infty} d\sigma \sigma^2 \eta_{\sigma_{\text{max}}}(\sigma) \int_0^{\pi} \left( \frac{2\pi d\theta_{\sigma} \sin\theta_{\sigma}}{(2\pi)^3} \right) \frac{2\pi}{\hbar} \\ &\times \left( v \frac{\hbar}{2M\omega(\boldsymbol{\sigma})} \frac{4}{9} \sigma^2 \left( \frac{\langle p_+^2 \rangle_{\text{Av}}}{2m} \right)^2 \right) \hbar\omega(\boldsymbol{\sigma}) \\ &\times \left[ (n(\boldsymbol{\sigma}) + 1) \delta \left( \frac{\hbar^2}{2m} 2k_+ \sigma \cos\theta_{\sigma} - \frac{\hbar^2 \sigma^2}{2m} - \hbar\omega(\boldsymbol{\sigma}) \right) \right. \\ &\left. - n(\boldsymbol{\sigma}) \delta \left( \frac{-\hbar^2}{2m} 2k_+ \sigma \cos\theta_{\sigma} - \frac{\hbar^2 \sigma^2}{2m} + \hbar\omega(\boldsymbol{\sigma}) \right) \right] \end{aligned} \quad (18)$$

and, carrying out the integration over  $\theta_{\sigma}$ ,

$$\begin{aligned} -\frac{d}{dt} \epsilon_+(\mathbf{k}_+) &= \frac{v(\langle p_+^2 \rangle_{\text{Av}}/2m)^2}{9\pi \hbar k_+} \frac{m}{M} \left[ \int_0^{\infty} d\sigma \sigma^3 \eta_{\sigma_{\text{max}}}(\sigma) \right. \\ &\left. \times \{ \eta_{2k_+ - \alpha}(\sigma) - n(\sigma) (\eta_{2k_+ + \alpha}(\sigma) - \eta_{|2k_+ - \alpha|}(\sigma)) \} \right], \end{aligned} \quad (19)$$

where  $\alpha = 2mv_{ac}/\hbar$ ,  $v_{ac} = \omega(\boldsymbol{\sigma})/\sigma$ , being the (longitudinal) acoustic wave velocity =  $1.8 \times 10^6$  cm/sec. for Au.

Equation (19) indicates that there are four distinct cases in the energy loss expression; I:  $2k_+ \geq \sigma_{\text{max}} + \alpha$ ; II:  $\sigma_{\text{max}} + \alpha > 2k_+ > \sigma_{\text{max}} - \alpha$ ; III:  $\sigma_{\text{max}} - \alpha \geq 2k_+ > \alpha$ ; IV:  $\alpha \geq 2k_+$ ; and, since  $\sigma_{\text{max}} \gg \alpha$ , these essentially reduce to three: I':  $k_+ > \sigma_{\text{max}}/2$ ; II':  $\sigma_{\text{max}}/2 \geq k_+ > mv_{ac}/\hbar$ . III':  $mv_{ac}/\hbar \geq k_+$ . Evaluating the integral over  $\sigma$  in Eq. (19) gives explicitly, with

$$A \equiv \frac{4}{9\pi} \hbar^{-1} \left( \frac{\langle p_+^2 \rangle_{\text{Av}}}{2m} \right)^2 \frac{m}{M} v(\sigma_{\text{max}}/2)^3,$$

$$-\frac{d}{dt} \epsilon_+(\mathbf{k}_+) = A \frac{\sigma_{\text{max}}}{2k_+}; \quad \hbar k_+ > \hbar \sigma_{\text{max}}/2, \quad (20\text{-I}')$$

$$\begin{aligned} -\frac{d}{dt} \epsilon_+(\mathbf{k}_+) &= A \left( \frac{2k_+}{\sigma_{\text{max}}} \right)^3 \left\{ \left( 1 - \frac{mv_{ac}}{\hbar k_+} \right)^4 \right. \\ &\left. - 8 \frac{mv_{ac}}{\hbar k_+} \left( \exp \left[ \frac{\hbar v_{ac} 2k_+}{\kappa T} \right] - 1 \right)^{-1} \right\}; \\ &\quad \hbar \sigma_{\text{max}}/2 \geq \hbar k_+ > mv_{ac}, \end{aligned} \quad (20\text{-II}')$$

$$+\frac{d}{dt} \epsilon_+(\mathbf{k}_+) = A \left( \frac{2mv_{ac}}{\hbar \sigma_{\text{max}}} \right)^3 8 \left( \exp \left[ \frac{2mv_{ac}^2}{\kappa T} \right] - 1 \right)^{-1}; \quad mv_{ac} \geq \hbar k_+, \quad (20\text{-III}')$$

where, for  $T \gg \hbar v_{ac} \sigma_{\text{max}}/\kappa \equiv T_{\text{Debye}} \gg 2mv_{ac}^2/\kappa$  one can expand both exponentials in the usual way.<sup>29</sup> The second inequality is easily satisfied in Au, and indeed in all metals. The thermalization time  $\tau$ , is now given by,

$$\begin{aligned} \tau &= \int_{mc^2/2 \cdot 137^2}^{\kappa T} \frac{d\epsilon_+}{[d\epsilon_+/dt]} \approx \frac{2}{Am\hbar\sigma_{\text{max}}} \int_{\hbar\sigma_{\text{max}}/2}^{mc/137} (\hbar k_+)^2 d(\hbar k_+) \\ &\quad + \left( \frac{\hbar^3 \sigma_{\text{max}}^3}{Am^8} \right) \int_{(2m\kappa T)^{\frac{1}{2}}}^{\hbar\sigma_{\text{max}}/2} \frac{d(\hbar k_+)}{(\hbar k_+)^2} \\ &= \frac{3}{2\pi} \frac{\hbar}{\langle p_+^2 \rangle_{\text{Av}}/m} \frac{M}{m} \frac{(\hbar\sigma_{\text{max}})^2}{\langle p_+^2 \rangle_{\text{Av}}} \frac{\hbar\sigma_{\text{max}}}{(2m\kappa T)^{\frac{1}{2}}} \\ &\quad \times \left[ 1 + \frac{(2m\kappa T)^{\frac{1}{2}}}{\hbar\sigma_{\text{max}}} \frac{8}{3} \left[ 2 \left( \frac{mc}{137\hbar\sigma_{\text{max}}} \right)^2 - 1 \right] \right] \\ &= 0.15 \times 10^{-10} \left[ \frac{mc^2/137^2}{\langle p_+^2 \rangle_{\text{Av}}/2m} \right]^2 \text{ sec.}, \end{aligned} \quad (21)$$

<sup>28</sup> As before,  $\eta_{x_0}(x) = 1$  if  $x < x_0$ ;  $\eta_{x_0}(x) = 0$  if  $x > x_0$ .

<sup>29</sup> Equations for the energy loss of an electron in an almost empty conduction band of an electrical non-conductor, entirely analogous to our Eqs. (20-I', II', III'), have been independently derived by Mr. W. R. Heller of this laboratory in the course of an investigation on the dielectric breakdown of non-polar insulators (to be published). *Note added in proof.*—F. Seitz (Phys. Rev. **76**, 1376 (1949)) has also independently derived essentially these equations (for the energy loss of an electron) in his work on electron multiplication in crystals.

where, to obtain the last expression, we have used  $T=300^\circ\text{K}$  and  $\hbar\sigma_{\text{max}}]_{\text{Au}}=0.8\text{ mc}/137$ . Then, with  $\langle p_+^2 \rangle_{\text{Au}}/2m]_{\text{Au}}\approx 0.22\text{ mc}^2/137^2$  (see below) we finally have<sup>30</sup>

$$\tau \approx 3 \times 10^{-10}\text{ sec.} \quad (22)$$

It may be mentioned that, according to Eq. (21),  $\tau$  is relatively insensitive to variations in temperature.

On the other hand, the mean life for slow positron annihilation, assuming that only the  $6s$  valence electrons are accessible to the positrons, is

$$\left\{ \frac{1}{v} \pi \left( \frac{e^2}{\text{mc}^2} \right)^2 c \right\}^{-1} = 25 \times 10^{-10}\text{ sec.} \approx 8\tau. \quad (23)$$

It is thus indeed quite probable that, as we have supposed throughout, the instant of annihilation will find the positron occupying states with  $\hbar k_+ \sim (2m\kappa T)^{\frac{1}{2}}$ ; such thermalized positrons possess therefore a total momentum very close to the bottom-of-the-band zero-point value, which, on a root-mean-square basis, is the  $\langle p_+^2 \rangle_{\text{Au}}^{\frac{1}{2}}$  used above.

It remains to justify the formula  $\langle p_+^2 \rangle_{\text{Au}}/2m]_{\text{Au}}\approx 0.22\text{ mc}^2/137^2$ . We have, with our choice of  $\psi_{\mathbf{k}+}(\mathbf{r})$  and with Eq. (9),

$$|\varphi_{\mathbf{k}+}(\mathbf{p}_+)|^2 = \text{const.} \sum_{\mathbf{g}_+} \left[ \delta_{\mathbf{g}_+,0} \left( 1 - \frac{2v_i}{v} \right) + \left( \frac{v_i}{v} \right)^2 \left\{ \left[ \sin(2\pi\mathbf{g}_+\mathbf{r}_i) - (2\pi\mathbf{g}_+\mathbf{r}_i) \cos(2\pi\mathbf{g}_+\mathbf{r}_i) \right]^2 \frac{9}{(2\pi\mathbf{g}_+\mathbf{r}_i)^6} \right\} \right] \delta(\mathbf{k}_+ + 2\pi\mathbf{g}_+ - \mathbf{p}_+). \quad (23)$$

Thus, recalling that for a face-centered cubic lattice (like Au) the  $\mathbf{g}_+$  of smallest magnitude are 0;

$$\pm a^{-1}(\mathbf{e}_1 + \mathbf{e}_2 - \mathbf{e}_3), \pm a^{-1}(\mathbf{e}_2 + \mathbf{e}_3 - \mathbf{e}_1), \pm a^{-1}(\mathbf{e}_3 + \mathbf{e}_1 - \mathbf{e}_2), \\ a^{-1}(\mathbf{e}_1 + \mathbf{e}_2 + \mathbf{e}_3); \pm 2a^{-1}\mathbf{e}_1, \pm 2a^{-1}\mathbf{e}_2, \pm 2a^{-1}\mathbf{e}_3;$$

etc. (here  $a$  is the lattice constant  $= (4v)^{\frac{1}{3}}$  and,  $\mathbf{e}_1, \mathbf{e}_2, \mathbf{e}_3$ , unit vectors along the cubic axes), one gets with  $v_i/v = \frac{1}{4}$ ,  $r_i = 1.0\text{ \AA}^\circ$ ,  $P(p_+)$ , the probability of the positron momentum having the

<sup>30</sup> From a calculation analogous to that in Eq. (21), the time required for the positron to slow down from a momentum

$$\frac{1}{3} \hbar\sigma_{\text{max}} \approx \frac{1}{4}(\text{mc}/137) \text{ for Au,}$$

to a momentum  $(2m\kappa T)^{\frac{1}{2}}$  is only 35 percent shorter than  $\tau$ . This justifies the statement made previously, in the discussion of the neglect of the collisions with  $2\pi\mathbf{g} \neq 0$ .

<sup>31</sup> The fact that one should use, in the Dirac formula for the annihilation mean life, the number of valence electrons per unit volume, and not the total number of electrons per unit volume, was first pointed out by Pomeranchuk, reference 2. It should, however, be mentioned that our estimate of the annihilation time may be too long by a factor of 2 to 4, because of the possibility of occasional annihilation against a  $5d$  electron and because of the neglect of the Coulomb attraction between the annihilating electron and positron (involving multiplication of the annihilation rate by

$$\sim \left\langle \frac{2\pi e^2 m}{\hbar |\mathbf{p}_- - \mathbf{p}_+|} \left( 1 - \exp \left[ -\frac{2\pi e^2 m}{\hbar |\mathbf{p}_- - \mathbf{p}_+|} \right] \right) \right\rangle_{\text{Av}}$$

We may also note that our time for annihilation is short compared to a reasonable estimate of the time required to trap the positron at a lattice defect (i.e. an  $\text{Au}^+$  vacancy, etc. For analogous estimates in the case of electron trapping, see N. F. Mott and R. W. Gurney, *Electronic Processes in Ionic Crystals* (Oxford University Press, London, 1940), p. 131 ff.).

value  $p_+^{\text{32}}$

$$P(p_+ = k_+) = \frac{|\varphi_{\mathbf{k}+}(p_+ = k_+)|^2}{\sum_{\mathbf{p}_+} |\varphi_{\mathbf{k}+}(\mathbf{p}_+)|^2} = 0.81, \\ P(p_+ = \left| \frac{2\pi\sqrt{3}}{a} \mathbf{g}_+ + \mathbf{k}_+ \right|) \approx \frac{8 |\varphi_{\mathbf{k}+}(p_+ = 2\pi\sqrt{3}a^{-1})|^2}{\sum_{\mathbf{p}_+} |\varphi_{\mathbf{k}+}(\mathbf{p}_+)|^2} = 0.14, \quad (24) \\ P(p_+ = \left| \frac{2\pi 2}{a} \mathbf{g}_+ + \mathbf{k}_+ \right|) \approx \frac{6 |\varphi_{\mathbf{k}+}(p_+ = 2\pi 2a^{-1})|^2}{\sum_{\mathbf{p}_+} |\varphi_{\mathbf{k}+}(\mathbf{p}_+)|^2} = 0.05, \\ P(p_+ \approx 2\pi g_+ > 2\pi 2/a) \approx 0.$$

Thus, the mean square positron momentum in the state  $\psi_{\mathbf{k}+}$  is

$$\langle p_+^2 \rangle_{\mathbf{k}+} \approx 0.81(\hbar k_+)^2 + 0.14(2\pi\sqrt{3}a^{-1})^2 + 0.05(2\pi 2a^{-1})^2, \quad (25)$$

and the mean square zero-point positron kinetic energy  $\langle p_+^2 \rangle_{\text{Au}}/2m$ , becomes, using  $a]_{\text{Au}} = 4.07\text{ \AA}^\circ = 7.7 \times 137\hbar/\text{mc}$ ,

$$\frac{\langle p_+^2 \rangle_{\text{Au}}}{2m} = \frac{\langle p_+^2 \rangle_{\mathbf{k}+}}{2m} - \frac{0.81(\hbar k_+)^2}{2m} \approx 0.22 \frac{\text{mc}^2}{137^2}. \quad (26)$$

We may conclude by noting that it is not quite correct, to assume that the electrons alone contribute to the center of mass momentum of the annihilating pairs and thus to picture the positrons as effectively stationary targets for annihilation.<sup>3</sup> This follows since, as we have just seen, even a thermalized positron has probabilities of the order of 14 percent and 5 percent, respectively, for possessing momenta as large as  $2\pi\sqrt{3}a^{-1} = 1.4\text{ mc}/137$  and  $2\pi 2a^{-1} = 1.6\text{ mc}/137$ . Indeed, our discussion makes use of no such assumption and obtains the probability amplitude for the momentum of the center of mass of the annihilating pairs, directly from the corresponding electron and positron coordinate space or momentum space wave functions (Eqs. (6), (7)).

## APPENDIX II

### The Annihilation Matrix Element

The integral of Eq. (6) for  $\Phi_{\mathbf{k}+, \mathbf{k}-}(\mathbf{p})$  is proportional to the matrix element of the annihilation transition in which an electron jumps from an originally occupied initial state with wave number  $\mathbf{k}_-$ , energy  $\approx mc^2$  to an originally unoccupied final state with wave number  $-\mathbf{k}_+$ , energy  $\approx -mc^2$ , with the simultaneous emission of two photons with total momentum  $\mathbf{p}$ . This can be straightforwardly shown from the usual two-step perturbation theory derivation of the annihilation probability based on the interaction of a Dirac electron with the quantized radiation field (see, e.g. W. Heitler, reference 6, pp. 204-208) provided that, (a): the wave functions of the intermediate (virtual) states of the process are treated in the free particle approximation, and (b): the wave functions of the initial and final states are treated non-relativistically, i.e. replaced by the corresponding Schrödinger wave functions ( $\psi_{\mathbf{k}-}(\mathbf{r}), \psi_{\mathbf{k}+}^*(\mathbf{r})$  which appear in Eq. (6)) times

$$\begin{pmatrix} 1 \\ 0 \\ 0 \\ 0 \end{pmatrix} \quad \text{and} \quad \begin{pmatrix} 0 \\ 0 \\ 1 \\ 0 \end{pmatrix},$$

respectively. Then, remembering that the electronic momenta involved in the intermediate states ( $\sim mc$ ) are much greater than those involved in the initial and final states ( $\sim mc/137$ ), and, using the orthonormality of the space parts of the intermediate state free particle wave functions, one can perform the sum over all the possible intermediate states and obtain Eq. (6) for  $\Phi_{\mathbf{k}+, \mathbf{k}-}(\mathbf{p})$ . Eq. (7) for  $\Phi_{\mathbf{k}+, \mathbf{k}-}(\mathbf{p})$  follows immediately from Eq. (6) by use of the Fourier transform relations between  $\psi_{\mathbf{k}\mp}(\mathbf{r})$  and  $\varphi_{\mathbf{k}\mp}(\mathbf{p}\mp)$ . It may also be mentioned that the above method yields most readily the known result<sup>19</sup> that the matrix element of the two-photon annihilation is proportional to the sine of the angle between the photon polarization vectors.

<sup>32</sup> We have used, in the first approximate equalities, the fact that for the positrons of interest,  $k_+ \approx \hbar^{-1}(2m\kappa T)^{\frac{1}{2}} \ll 2\pi\sqrt{3}a^{-1}$ , etc.