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On the breadths of annihilation lines in copper and in gold

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Summary. The breadths of annihilation lines produced in the annihilation process of electrons and positrons are calculated for copper and gold. Two models are considered: 1^o The Fermi gas model of conduction electrons and Cu^+ or Au^+ ions and 2^o The model where the atoms exist complete in the crystal, but where, however, owing to the influence of the crystal the outer shell 4 s of Cu is so much compressed that it overlaps with the 3d shell (and for Au the 6 s shell overlaps with the 5 d shell). The breadths due to conducting electrons and the electrons in different shells of the Cu and Au ions are calculated. By comparison of the results with the experimental data it is shown that in the first model the contribution of conducting electrons to the breadth of the annihilation line is negligible. For both models the largest contribution stems from the most external complete shell but in Cu about 5% and in Au about 40–50% of the annihilations take place in the next inner shells of the atoms.

§ 1. Introduction

The purpose of the present paper is the theoretical investigation of the shape of the lines of γ radiation produced in the process of annihilation of positrons for the examples copper and gold. Precision measurements of the shape of the annihilation line in copper were made by DU MOND, LIND and WATSON (DU MOND 1954) with a crystal spectrometer. They used an activated copper sample in which β -radioactive Cu^{64} atoms were present. The positrons produced in β decay (having a maximum initial energy of 0,66 MeV) annihilate with the electrons in the sample giving rise to the annihilation line.

According to HEITLER (1954 p. 270) the probability of annihilation of fast positrons is small, so that 98% of the positrons are stopped (or slowed down to thermal velocities, which we neglect) by collisions with the electrostatic fields of nuclei before being annihilated. These positrons annihilate then with the slow conduction electrons or with electrons of different atomic shells. The center of mass of the annihilating pair moves with the velocity of half the velocity of the electron, so that in the laboratory system the annihilation photons have slightly different wave lengths

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and therefore the annihilation line exhibits a certain structure owing to Doppler broadenings of different orders of magnitude. DU MOND and al. also made a rough estimate of these Doppler broadenings to annihilations in different electron shells.

Since the annihilating electrons are moving, the two annihilation photons are emitted at angles slightly different from π . DE BENEDETTI, COWAN, KONNECKER, and PRIMAKOFF (DE BENEDETTI 1949) have measured the departure from antiparallelism of the two photons in the process of pair annihilation in gold and have, therefrom calculated the mean momentum of the center of mass of the annihilating pair.

The main result of both these papers was the statement that almost all the positrons are slowed down to thermal velocities in the sample and then annihilate with the conduction electrons.

In this paper two extreme models are considered. In the first the metal consists of Cu^+ or Au^+ ions and a degenerate Fermi gas containing the conduction electrons. In the second model atoms exist complete but, however, according to KRUTTER (1935) owing to the influence of the crystal the outer 4 s shell for Cu is so much compressed that it overlaps with the 3 d shell (and for Au, the 6 s shell overlaps with the 5 d shell). In both cases the positrons are assumed to be stopped before annihilation. In the process of annihilation both the electron and the positron may be regarded as free particles; since they have equal and opposite charges the electrostatic potential of the nuclear field has no influence on the energies of the annihilation photons. We neglect the cases when positronium is formed before annihilation.

In § 2 we calculate the annihilation rate of a moving free electron annihilating with a positron at rest. Afterwards (§ 3) we expand the wave function of an atomic electron into plane waves and calculate the rate of annihilation of the positron at rest with the electron having this wave function. In § 4 and 5 we apply the formulae of § 3 to calculate the annihilation rates for different shells and the conducting electrons in copper and gold and compare them with experimental data. § 6 contains the results and discussion.

§ 2. Annihilation of a free electron and a free positron in the system where the positron is at rest

We consider a free electron with momentum \mathbf{p} and energy E annihilating with a positron at rest. The two emitted annihilation photons have momenta \mathbf{k}_1 and \mathbf{k}_2 . Since both particles considered are free, energy and momentum are conserved

$$\mathbf{p} = \mathbf{k}_1 + \mathbf{k}_2 \quad (1)$$

$$E + \mu = k_1 + k_2, \quad E = \sqrt{p^2 + \mu^2} \quad (2)$$

The transition probability from the initial state 0 (electron \mathbf{p} , a hole at rest) to the final state F is derived from the compound matrix element K_{FO} . Using standard methods and the notation given by HEITLER (1954, p. 213–219) this is given by

$$K_{FO} = \frac{e^2 (2\pi \hbar^2 c^2)}{2\mu \sqrt{k_1 k_2}} (u_F^* \{ \alpha_1 \alpha_{k_1} \alpha_2 + \alpha_2 \alpha_{k_2} \alpha_1 + 2 \mathbf{e}_1 \cdot \mathbf{e}_2 \} u_0)$$

where

$$\alpha_{k_1} \equiv \frac{\boldsymbol{\alpha} \cdot \mathbf{k}_1}{k_1} \quad \text{etc.}$$

As we are not interested in the probability of annihilation of an electron and a positron with definite spins we average over their spin directions. The transition probability to the final state with definite directions of polarisation of both annihilation photons is given by

$$w = \frac{2\pi}{\hbar} \left(\frac{1}{4} S_F S_0 |K_{FO}|^2 \right) \varrho_F$$

where ϱ_F denotes the number of final states per energy intervall dE_F . The final state is completely determined by k_1 and the angle β between \mathbf{k}_1 and \mathbf{p} . Therefore we have

$$\varrho_F dE_F = \varrho_{k_1} dk_1, \quad \varrho_{k_1} = \frac{d\Omega k_1^2}{(2\pi \hbar c)^3} \quad (3)$$

From the conservation laws (1) and (2) we have

$$k_1 (\mu + E - p \cos \beta) = \mu (\mu + E) = \mu (k_1 + k_2) \quad (4)$$

and

$$E_F = k_1 + k_2 = k_1 + \sqrt{p^2 - 2k_1 p \cos \beta + k_1^2}$$

so that

$$\left(\frac{\partial k_1}{\partial E_F} \right)_\beta = \frac{k_1 k_2}{\mu (k_1 + k_2)} \quad (5)$$

Hence from (3) and (5):

$$\varrho_F = \varrho_{k_1} \left(\frac{\partial k_1}{\partial E_F} \right)_\beta = \frac{1}{(2\pi \hbar c)^3} \frac{k_1^3 k_2}{\mu (k_1 + k_2)} d\Omega$$

Applying the usual methods of evaluating the S-sums we obtain ($r_0 = e^2/\mu$)

$$w = \frac{c r_0^2 d\Omega k_1^2}{8 E (k_1 + k_2)} \left(-4 \cos^2 \Theta + \frac{k_1}{k_2} + \frac{k_2}{k_1} \right) \quad (6)$$

where Θ is the angle between \mathbf{e} , and \mathbf{e}_2 . If there are N electrons and N_+ positrons in the cube (of volume 1 cm^3) the rate of annihilation is

$$dR' = \frac{N N_+ c r_0^2 d\Omega k_1^2}{8 E (k_1 + k_2)} \left(-4 \cos^2 \Theta + \frac{k_1}{k_2} + \frac{k_2}{k_1} \right) \quad (7)$$

As we do not want to deal with polarized photons we shall sum (7) over both polarizations of both photons. The annihilation rate into unpolarized photons is

$$dR'' = \frac{1}{2E} N N_+ c r_0^2 d\Omega \frac{k_1^2}{k_1 + k_2} \left(\frac{k_1}{k_2} + \frac{k_2}{k_1} + \sin^2 o \right)$$

where o is the angle between k_1 and k_2 .

Because of the conservation laws (1) and (2) k_1 , k_2 , E and o are not independent. We can express, therefore, the annihilation rate as a function of $P \equiv p/\mu$ and $K \equiv k_1/\mu$ only:

$$dR'' = \frac{N N_+ c r_0^2 d\Omega K^2}{2 (\sqrt{1+P^2+1}) \sqrt{P^2+1}} \left\{ \frac{K^2+2K-2}{\sqrt{1+P^2+1}-K} + \frac{\sqrt{P^2+1}+3-K}{K} - \frac{1}{K^2} - \frac{1}{(\sqrt{1+P^2+1}-K)^2} \right\} \quad (8)$$

We shall call K and P the reduced momenta.

Owing to the conservation laws, K can, for a given P , only vary within certain limits. Since in formula (4) the angle β is arbitrary we have

$$K_{min} \equiv \frac{1 + \sqrt{P^2+1}}{1 + \sqrt{P^2+1} + P} \leq K \leq \frac{1 + \sqrt{P^2+1}}{1 + \sqrt{P^2+1} - P} \equiv K_{max} \quad (9)$$

For $P = 0$ we have $K = 1$ only, for $P \rightarrow \infty$

$K_{min} \rightarrow 1/2$, $K_{max} \rightarrow \infty$ (see Fig. 1).

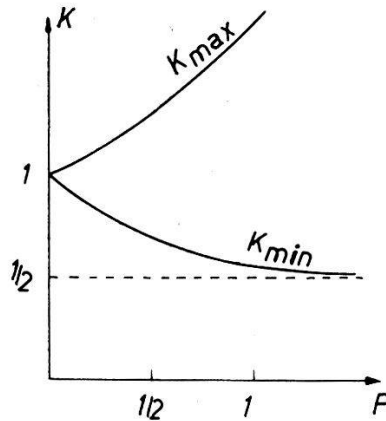


Fig. 1

§ 3. Annihilation of a positron at rest with a bound electron

Since the electron and positron have equal and opposite charges the electrostatic potential of the nucleus plays no rôle in the annihilation process and we can treat the particles as free. In order to calculate the annihilation rate with a bound electron we expand the wave function of the electron into plane waves and apply formula (8) to each component.

In the non-relativistic (N. R.) approximation the expansion into plane waves is

$$\psi_{Dirac}(\mathbf{r}, t) = \int \frac{d^3p}{(2\pi\hbar c)^{3/2}} u e^{\frac{c}{\hbar} \left(\frac{1}{c} \mathbf{p} \cdot \mathbf{r} - Et \right)} C(\mathbf{p}) \quad (10)$$

where u is the spinor, $u = \begin{pmatrix} 1 \\ 0 \\ 0 \\ 0 \end{pmatrix}$

and

$$C(\mathbf{p}) = \frac{1}{(2\mu\hbar c)^{3/2}} \int \psi_{Schröd.}(\mathbf{r}, t) e^{-i \frac{\mathbf{p} \cdot \mathbf{r}}{\hbar c}} d^3x \quad (10')$$

$C(\mathbf{p})$ is normalized.

The probability for the electron to have the momentum between \mathbf{p} and $\mathbf{p} + d\mathbf{p}$ is $|C(\mathbf{p})|^2 d^3p$ and the number of electrons having the reduced momenta between \mathbf{P} and $\mathbf{P} + d\mathbf{P}$ is

$$N |C(\mathbf{P})|^2 d^3P \quad (11)$$

where

$$C(\mathbf{P}) = \mu^{3/2} C(\mathbf{p}/\mu)$$

The annihilation rate coming from electrons with reduced momenta between \mathbf{P} and $\mathbf{P} + d\mathbf{P}$ is (from (8) and (11)) in the N. R. approximation ($P^2 \ll 1$): equal to

$$dR'' = \frac{1}{4} N N_+ c r_0^2 d\Omega K^2 \left\{ \frac{K^2 - 2K - 2}{2 - K} + \frac{4 - K}{K} - \frac{1}{K^2} - \frac{1}{(2 - K)^2} \right\} |C(\mathbf{P})|^2 d^3P \quad (12)$$

where K fulfills the inequalities

$$\frac{1}{1 + \frac{P}{2}} \leq K \leq \frac{1}{1 - \frac{P}{2}} \quad (13)$$

The total annihilation rate for a given K is obtained by integrating (12) over all possible P corresponding to this K i.e. from $P_{min} = 2 |K - 1|/K$ to infinity as follows from (13) (see Fig. 1). Actually it is not quite correct to integrate over P to infinity because we use the non relativistic approximation, but for the cases considered in this paper $|C(\mathbf{P})|^2$ vanishes so quickly that we do not make a considerable error. We have, therefore, the annihilation rate

$$R = \frac{1}{4} N N_+ c r_0^2 d\Omega K^2 \left\{ \frac{K^2 - 2K - 2}{2 - K} + \frac{4 - K}{K} - \frac{1}{K^2} - \frac{1}{(2 - K)^2} \right\} \int_{2|K-1|/K}^{\infty} |C(\mathbf{P})|^2 d^3P \quad (14)$$

§ 4. Annihilation in copper

We now consider the case of copper. We consider the two limiting models mentioned in the introduction.

1. The Fermi-gas model. The metal is composed of Cu^+ ions and conducting electrons forming a totally degenerate Fermigas and possessing therefore the distribution function

$$C(P) = \begin{cases} 1 & \text{for } 0 \leq P \leq P' \\ 0 & \text{for } P > P' \end{cases} \quad (15)$$

From the value 7.04 eV for the maximum energy of the conduction electrons in copper at zero temperature (SEITZ, 1943, p. 146) we obtain the value of $P' = 1.4 \cdot 10^{-5}$.

2. We consider Cu atoms forming a lattice whose filled shells are approximately the same as in free Cu atoms and the wave function of the 4 s electrons overlaps strongly with that of the 3 d shell (KRUTTER 1935).

Since the maximum energy of positrons emitted from the radioactive Cu^{64} atoms is equal to 0.66 MeV, they can penetrate deeply into the cores of the atoms and we cannot exclude a priori the possibility of annihilation also in the inner shells.

In the Fermi-gas model the integral (14) becomes for the conduction electrons:

$$J_c \equiv \int_{\frac{2|K-1|}{K}}^{\infty} |C(P)|^2 d^3P = \frac{4\pi}{3} P'^3 \left[1 - \left(\frac{2}{P'} \frac{|K-1|}{K} \right)^3 \right]$$

Expressing K in terms of the wave length λ in X-units

$$\lambda = \frac{\lambda_0}{K} \quad (16)$$

where $\lambda_0 = 24.27$ \AA is the Compton wave length of the electron. We have

$$J_c = \frac{4\pi}{3} P'^3 \{ 1 - (15.5 |\Delta\lambda|)^3 \}, \quad \Delta\lambda \equiv \lambda - \lambda_0 \quad (17)$$

For the Cu^+ ions we consider the HARTREE diagram for the charge density (HARTREE 1933, p. 300)²⁾. This function has three peaks corresponding to the three shells: K (2 electrons), L (8 electrons) and M (18 electrons) at distances $r_{n \max}$ from the nucleus:

$$\begin{aligned} r_{1 \max} &= 0.035 \cdot 10^{-8} \text{ cm} \\ r_{2 \max} &= 0.069 \cdot 10^{-8} \text{ cm} \\ r_{3 \max} &= 0.32 \cdot 10^{-8} \text{ cm} \end{aligned} \quad (18)$$

²⁾ In the calculations leading to fig. 2 the exchange effect has not been taken into account, but the corrections due to exchange are in our case negligible.

For simplicity we replace the two first peaks by one placed at

$$(2 r_{1 \max} + 8 r_{2 \max})/10 = 0,062 \cdot 10^{-8} \text{ cm} \approx r_{2 \max} \quad (18')$$

We replace now the wave function for each shell by the normalized hydrogen-like ground state wave function ψ_n with parameters so chosen that $r^2 |\psi_n|^2$ has the maximum at $r_{n \max}$:

$$\psi_n = a_n^{3/2} \pi^{-1/2} e^{-a_n r} \quad (19)$$

where

$$a_n = 1/r_{n \max}$$

Then $r^2 |\psi_n|^2$ approximates roughly the charge density function for the corresponding shell. Since the regions where the corresponding density functions are appreciably different from zero overlap only to a small degree we shall treat the annihilation in different shells as separate phenomena. Inserting (19) into (10) we have

$$C_n(\mathbf{P}) = -\frac{4}{\pi \sqrt{2}} \frac{(a_n \hbar c)^{5/2}}{[(a_n \hbar c)^2 + P^2]^2}$$

and from (11⁰)

$$C_n(\mathbf{P}) = -\frac{4}{\pi \sqrt{2}} \frac{A_n^{5/2}}{(P^2 + A_n^2)^2} \quad (20)$$

where

$$A_n = \frac{a_n \hbar c}{\mu} = a_n \lambda_0 = 3,86 \cdot 10^{-11} a_n \quad (21)$$

(λ_0 is $(1/2\pi)$. Compton wave length for the electron).

From (18), (18') and (21) we get

$$A_2 = 0,062 \quad A_3 = 0,012 \quad (22)$$

It is easily seen that

$$A_n = \sqrt{P^2} = \sqrt{\beta_n^2}$$

where $\sqrt{\beta_n^2}$ are the mean velocities of the electrons in different shells and they have the same orders of magnitude as those given by DU MOND (1946, p. 1237).

From (14) and (20) we get the annihilation rate in the n shell putting in $\{\}, K=1$ because of smallness of A_n , (η denotes the relative probability of annihilation in the n shell)

$$dR_n = c N N_+ r_0^2 d\Omega \frac{16}{\pi} \eta_n \int_{2|K-1|/K}^{\infty} \frac{A_n^5 P^2 dP}{(P^2 + A_n^2)^4} \quad (23)$$

Evaluating the integral and expressing K in terms of the wave lengths λ we have

$$dR_n = c N N_+ r_0^2 d\Omega \frac{16}{\pi} \eta_n J_n \quad (24)$$

where

$$J_n = \frac{1}{16} \left(\frac{\pi}{2} - \operatorname{arctg} \frac{2|\Delta\lambda|}{\lambda_0 A_n} \right) + \frac{\Delta\lambda}{\lambda_0 A_n} Y \left(\frac{1}{3} Y^2 - \frac{1}{12} Y - \frac{1}{18} \right) \quad (24')$$

$$Y = \frac{(\lambda_0 A_n)^2}{(2\Delta\lambda)^2 + (\lambda_0 A_n)^2}$$

In Fig. 2 dR_n are drawn as a function of $\Delta\lambda$. Since the dR_n are practically symmetrical only the left parts of them are drawn. In this figure also the experimental curve of DU MOND and al. (1949) corresponding to the GAUSSIAN curve $C \exp(-x^2/2\sigma)$ for $\sigma = 0.096$ X. U.

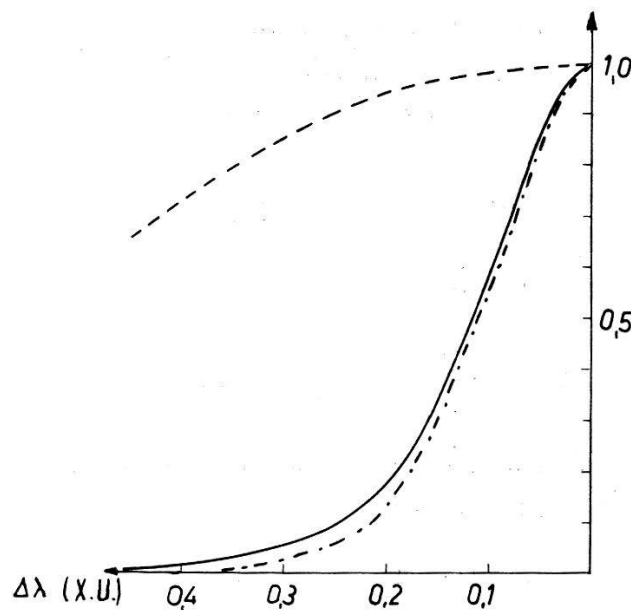


Fig. 2

- exp. curve of du Mond
- - - theor. curve for K and L shells
- · - theor. curve for M shell

All curves are normalized so, that their ordinates for $\Delta\lambda = 0$ are equal to 1.

The curve (17) for the conduction electrons is not drawn because it has an extremely small breadth in comparison with the dR_m -curves. From Fig. 3 and from (17) one can easily state that the main contribution to annihilation comes from the 18 electrons of the M shell, the contribution of the conduction electrons is negligible and the contribution of the K and L shells does not exceed 5%. The total annihilation rate is therefore

$$dR = 0,05 dR_2 + 0,95 dR_3$$

(For the electrons of velocity zero we get, after integration over angles, the annihilation rate

$$R = 2 \pi c N N_+ r_0^2$$

in agreement with Heitler's formula (1954, § 27 (11)³).

In the second model we consider the 4 s electrons as belonging to the *M* shell. From the preceding calculations we can state that according to this model the contribution of the *M* shell together with the 4 s electrons amount to 95% and the contribution from the *K* and *L* shells to 5%.

§ 5. Annihilation in gold

The measurements in gold were made by DE BENEDETTI and al. (1950). In their experiment a radioactive copper source emitted positrons with maximum energy 0,66 MeV which entered into the sample of gold and annihilated with the electrons inside this sample. DE BENEDETTI and al. observed the angular correlations of the photons of annihilation and from their experiments they deduced that the mean momentum \bar{p}_{Au} of the center of mass of annihilating particles was 1.2/137 mc or in our notation

$$\bar{p}_{m.c.} = \frac{1,2}{137} = 0,088 \quad (25)$$

In order to apply our method to this case we must make some introductory remarks.

1. If we compare the GAUSSIAN curve $f(x) = C \exp(-x^2/2\sigma^2)$ with the curve (24) with the same height and the same half breadth we may easily deduce the relation between σ and A :

$$A = 0,13 \sigma \quad (26)$$

2. For a GAUSSIAN curve $f(x)$ the mean deviation from λ_0 is equal to σ

$$\overline{\Delta\lambda} = \sqrt{(\lambda - \lambda_0)^2} = \sigma \quad (27)$$

3. The deviation $\Delta\lambda$ of λ from 24, 27 X. U. is the result of the Doppler effect since the two annihilating photons are emitted from the moving center of mass with the mean velocity v :

$$\frac{v}{c} = \frac{|\overline{\Delta\lambda}|}{\lambda_0}$$

To get the momentum of the centre of mass we multiply this formula by the mass of the mass centre and get, remembering (27)

$$\bar{P}_{m.c.} = 2 \frac{6}{\lambda_0} = 0,082 \sigma$$

³) Our result differs from HEITLER's formule by the factor $2 N_+$ since HEITLER defines $R = 2 N \Phi v_+$ and in our paper $R = 2 N N_+ \Phi$ · relative velocity of electron and positron.

(For Cu, $P_{m.c} = 1.1/137$, (in agreement with DE BENEDETTI). For Au we have from (25) and (28)

$$\sigma_{exp} = 0.107 \text{ X. U.}$$

Since for Au from experiment only $P_{m.c}$ is given and not the whole shape of the annihilation line we shall approximate the experimental and theoretical curves by GAUSSIAN curves.

We regard for gold the same two models as for copper. The maximum energy of conduction electrons in the Fermi-gas model is according to SEITZ (1940) equal to 5.04 eV and therefore the line breadth is even smaller than in the case of copper. In the second model we assume in analogy to the case of copper that the wave function of the 6 s electrons overlaps strongly with that of the 5d electrons.

The theoretical $r_{n \max}$ for $n = 4, 5$ were computed from the paper of Henry on the Hartree model for Au^+ (HENRY 1954).

$$r_{4 \max} = 0.22 \cdot 10^{-8} \text{ cm}$$

$$r_{5 \max} = 0.5 \cdot 10^{-8} \text{ cm}$$

From (27) and (21) we have at once

$$A_4 = 0.0175 \quad \sigma_4 = 0.135$$

$$A_5 = 0.0078 \quad \sigma_5 = 0.059$$

The corresponding GAUSSIAN curves are drawn in Fig. 3 (left parts). The normalization of the curves is the same as in Fig. 2.

In the first model we may interpret the dotted curve which agrees best with the experimental one as the curve for the contribution of the 50–60% of annihilations in the *O*-shell and a contribution of 40–50% of annihilation in the *N*-shell. The contribution of conduction electrons is negligible. In the second model we may interpret 50–60% of the annihilations as coming from the 18 electrons of the *O*-shell together with the one 6 s electron.

DE BENEDETTI and al. used the intermediate model in which the 6 s electrons have the same wave functions as the 5 d electrons when they are inside the ion and are free when they are outside it. The positrons are assumed to be stopped and annihilated outside the ions. DE BENEDETTI and al. obtained 3/4 of the effect from this model, which is in qualitative agreement with the curve for conduction electrons in fig. 3. But fig. 3 indicates that almost the whole contribution to annihilation stems from the electrons inside the ions.

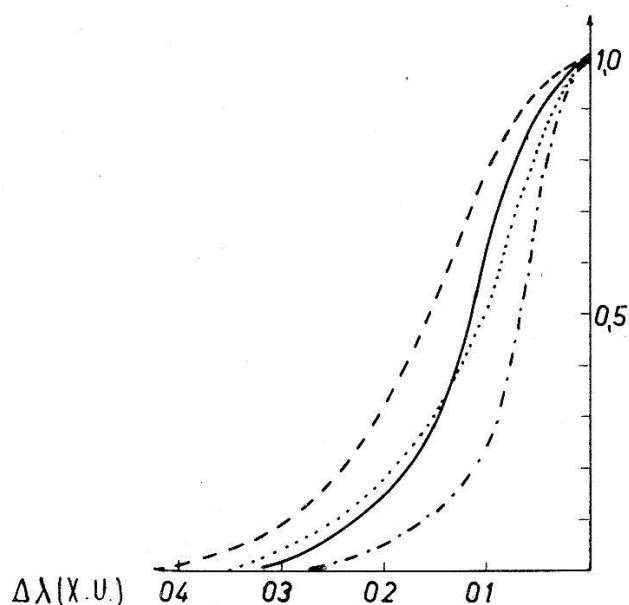


Fig. 3

- curve from exp. datum
- theor. curve for N shell
- theor. curve for O shell
- resultant theor. curve

§ 6. Results and discussion

From the comparison of the theoretical curves for different shells and the experimental data for Cu and Au discussed in the preceeding sections it follows for the Fermi-gas model that the contribution from conduction electrons to the annihilation line breadth is negligible and the greatest contribution (95% for Cu, and 50–60% for Au) stems from the annihilation in the outmost closed shell of the atom. But some of the positrons (5% in Cu, 40–50% in Au) are stopped and annihilate before reaching thermal velocities.

In the second model the contribution of the outside shell together with the last incomplete shell (one electron) is for Cu about 95%, for Au 50–60%. The remaining contributions are due to the next inner shell. We compare now our results with the estimations of DU MOND and al. We see from (20) and (22) that we obtain for the M shell the theoretical value $\sigma_3 = 0.094$ X. U., whereas from the half breadth value of 0.21 X. U. given by DU MOND it follows for the corresponding GAUSSIAN curve $\sigma = 0.09$ X. U. These two values agree fairly well. We now repeat the method of estimation of DU MOND and al. for the conduction electrons in Au. The mean energy of these electrons is 3.5 eV. The mean velocity of the mass centre of annihilating particles is 0.0012 c and according to (27) and (27') we get $\sigma = 0.008$ X. U. This value is much greater than the exact value following from (17') but is ten times smaller than the value

estimated (in this case not quite correctly) by DU MOND and al., as coinciding nearly with the experimental value $\sigma = 0.096 \text{ X. U.}$

If our simple picture of the mechanism of annihilation is true, the annihilation line should be accompanied for copper by the M spectrum of characteristic X-rays and for gold by the characteristic N -spectrum (independently of the assumption of the first or the second model).

I wish to express my thanks to Professor W. HEITLER for his kind interest in this work and helpful discussions. I also thank Dr. Z. NARAY and Dr. L. O'RAIFEARTAIGH for interesting discussions, the Schweizerischer Nationalfonds for the grant enabling me to stay in Zurich and the Polish Ministry of Higher Education for the granting of leave of absence.

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