On the Spatial Distribution of Photoelectrons Ejected from the Atomic X-Shell

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With an electron velocity analyzer and modified Geiger-Mueller electron counter, the longitudinal space-distribution of electrons ejected from the K energy level of copper by Mo $K\alpha_1$ has been determined. Asymmetries in the experimental and theoretical distributions are compared and lack of satisfactory accord with wave mechanics is found. The effects of nuclear scattering are briefly discussed. A convenient asymmetry factor, k , is introduced. It is possible to compute k from data given by other

I. INTRODUCTION

N 1927, Auger and Perrin' and Wentzel' announced, respectively, their "classical" and wave-mechanical expressions for the distribution in space of x-ray photoelectrons. Since then, several experimental studies^{3, 4, 5, 6} on photoelectrons produced in gases have been carried out. For the most part, the investigations have been on the longitudinal distribution of electrons ejected from the K -shells of the heavier atoms or from the lighter atoms where the binding energy was negligible. Auger³ and Anderson⁵ succeeded in studying the fog-tracks of electrons from the L -shells and agreed in the important finding that the distribution curves for these electrons are markedly more isotropic than those for the Xelectrons. Watson and Van den Akker^{7, 8} found that the "spread" in the distribution of electrons ejected from very thin metallic films is definitely investigators and, from an analysis of the statistical errors in the C. T. R. Wilson method, to compute probable errors. This is done and it is shown that a probably real difference, amounting to 10 percent, exists between the experimental and wave-mechanical asymmetries. Computations from an early but erroneous wave-mechanical treatment are in excellent accord with experimental. results.

a function of the type of level from which the electron is ejected, as well as of the binding energy and the frequency of the incident x-rays. As regards comparison of experiment with theory, we may say that the distribution of L-electrons is only in qualitative agreement with wave-mechanics, while the distribution for Xelectrons seems generally to be regarded as being in agreement with theory. Sommerfeld and Schur⁹ and Auger and Miss Meyer³ have noted, however, that the asymmetry in the experimental distributions for K -electrons seems to be in better agreement with an early but erroneous wave-mechanical treatment which has since been corrected.⁹ It is the purpose of this paper to present new results on the longitudinal distribution of electrons ejected from the K -shell of atoms composing a very thin metallic film and to analyze results obtained in the past by other investigators who used the C. T. R. Wilson method. There appears to be a real discrepancy between experiment and present wave-mechanical theory.

II. APPARATUS

Photoelectrons ejected from the X-shell of copper by Mo $K\alpha_1$ were counted with a modified Geiger-Mueller tube' after they had been sepa-

¹ P. Auger and F. Perrin, J. de Physique 8, 93 (1927).

² G. Wentzel, Zeits. f. Physik 41, 828 (1927).

³ P. Auger, C. R. 186, 758 (1928); J. de Physique 9, 225 (1928);C. R. 187, 1141 {1928);C. R. 188, 447 (1929);C. R. 188, 1287 (1929); P. Auger and Miss Meyer, C. R. 192, 672 $(1931).$

E.J. Williams, J. M. Nuttall and H. S. Barlow, Proc. Roy. Soc. A121, 611 (1928).

C. D. Anderson, Phys. Rev. 35, 1139 (1930).

^{&#}x27;E. Lutze, Ann. d. Physik (5) 9, ⁸⁵³ (1931).

⁷ E. C. Watson and J. A. Van den Akker, Proc. Roy. Soc. A126, 138 (1929).

⁸ J. A. Van den Akker and E. C. Watson, Phys. Rev. 37, 1631 (1931).

⁹A. Sommerfeld and G. Schur, Ann. d. Physik (5) 4, 409 {1930).

rated from other electrons by an electrostatic refocusing analyzer of the type devised by Hughes and Rojansky¹⁰ and Hughes and Mc-Hughes and Rojansky¹⁰ and Hughes and Mc
Millen.¹¹ The copper film, deposited on cello phane by evaporation, was approximately 130 atoms thick and 0.23 cm wide. This film was mounted at the center of an evacuated brass chamber, the latter being capable of rotation and having 16 ports for the admission of the x-ray beam so that θ (the angle between the forward direction of the x-ray beam, and the initial direction of ejection) could be varied in 10' steps from 10° to 170° , 90° excluded. To reduce scattering to a minimum, the copper film was oriented so that the initial direction of all electrons received by the analyzer was normal to the surface of the copper film. Since the entrance slit of the analyzer was 2.85 cm from the film, the uncertainty produced in θ by the width of the film was only 2.3° (a negligible figure in the longitudinal distribution). The entrance and exit slits of the analyzer were 0.031 cm wide and the radii of the cylindrical analyzer plates were 4.85 cm and 5.15 cm. These dimensions ensure good resolution, which made it possible to resolve the Mo K_{α_1} : Cu K-electrons from the Mo K_{α_2} : Cu K, and Mo K_{α_1} : Cu K-electron which had suffered energy losses in escaping from the film. The x-ray beam, generated in an trom the film. The x-ray beam, generated in an
oil-immersion balanced-circuit outfit,¹² was sufficiently wide to include the copper film at all angular settings.

III. METHQD AND REsULTs

Since the electron currents were, at maximum, less than 5 electrons per minute, it seems desirable to outline the method of measurement. Happily, one electron-counter served throughout the period of nearly three months required for the results given in this paper, without suffering change of sensitivity or exhibiting erratic bechange ot sensitivity or exhibiting erratic be
havior.¹³ The residual count (analyzer voltag

zero, x-rays on or off) was only 1.6 ions per minute. The residual count was not usually taken but, rather, the procedure was to take electron counts over the Mo K_{α_1} : Cu K peak and at the high energy foot of the peak (the latter being the true background count). All counts were taken over ten-minute intervals, alternating from the peak to the foot of the energy distribution curve, until eleven tenminute counts had been recorded for the peak and an equal number for the foot. All counts were reduced to number per minute. The difference between the average number of impulses (Geiger-Mueller) per minute at peak (P) and foot (F) was taken as a measure of the electron current at the given angular setting. In a typical case, the average number of impulses at peak and foot were, respectively, 7.46/min. and 3.30/min. Thus, a period of four hours of continuous observation was required for the measurement of a current of about 4.2 electrons/min. In general, several of these four-hour determinations were made to establish the current at a given angle. The results of 69 four-hour determinations are shown in Fig. 1.

Fro. 1. Of statistical interest, this figure gives individual determinations of electron currents. Each of the 69 determinations results from 22 ten-minute counting periods over the peak and foot of the energy-distribution curve at the given angular setting.

Averages of the points shown in Fig. 1 are given in Fig. 2, with twice the probable errors

¹⁰ A. L. Hughes and V. Rojansky, Phys. Rev. 34, 284 $(1929).$

¹¹ A. L. Hughes and J. H. McMillen, Phys. Rev. 34, 291 (1929).

¹² R. D. Bennett, N. S. Gingrich and W. C. Pierce, Rev. Sci. Inst. 2, 226 (1930).

 13 It is perhaps incorrect to speak of the "sensitivity" of a Geiger-Mueller tube, because this device records the

formation of individual ions; when imperfect, spurious impulses are recorded in such a way that the "sensitivity" apparently increases.

FrG. 2, Experimental and theoretical distributions, giving relative probabilities of ejection per unit solid angle. Dashed curve, experimental; full curve, wave-mechanics; dot-dash curve, classical.

represented by the vertical lines drawn through the points. The probable errors were computed from the relation, $r = 0.67[(P+F)/(10\times11 m)]^{\frac{1}{2}}$ where P and F are defined as above and m is the number of four-hour determinations at the angle in question. (This relation is essentially the square root of the sum of the squares of the probable errors in P and F .)

Determination of the longitudinal distribution by the C. T. R. Wilson method involves counting the number of fog-tracks occurring in a given angular range and hence, by this method, the probability per unit angle, $P(\theta)$, is found. Since the solid angle for a given angular range tends to go to zero at $\theta = 0^{\circ}$ and $\theta = 180^{\circ}$, actually few electrons are counted at these extreme angles and consequently little weight can be attached to the corresponding experimental values of $P(\theta)$. It therefore seems probable that effects due to weak scattering would not be noticeable where this method is used. In the present work, on the other hand, the electrons were received by a slit and hence the probability per unit solid angle, $F(\theta)$, was determined directly. Electron counts taken near 0' and 180', therefore, possess considerable weight. Inspection of Fig. 2 shows that the electron currents at 0° and 180° do not. go to zero; it cannot be said, at the present time, whether the major part of these currents are real or due to the spurious action of nuclear scattering.

FIG. 3.Experimental and theoretical distributions, giving relative probabilities per unit angle; the ordinates of this figure are those of Fig. 2 multiplied by $\sin \theta$.

The solid curve in Fig. 2 represents the wavemechanical expression for K -electrons ejected by mechanical expression for
unpolarized rays,^{14, 15, 16, 17}

$$
F(\theta) \propto \frac{\sin^2 \theta}{\left[1 - (\beta/a) \cos \theta\right]^4}
$$

adjusted to fit the experimental points at the peak. In this expression β is the ratio of the speed of the photoelectron to that of light and $a = 1 + \alpha/2 = 1 + h\nu/2mc^2$, where v is the frequency of the incident x-rays. The classical expression given by Auger and Perrin' is giv'en by the dash-dot curve.

For the comparison of experiment with theory, it is convenient (for mathematical and graphical reasons) to convert the experimental results and theory into the form $P(\theta)$. This is simply effected by multiplying all experimental and theoretical ordinates in Fig. 2 by sin θ . The result of doing this is shown in Fig. 3, in which the ordinates express the probability per unit angle; and, again, twice the probable errors are given by the vertical dashes through the points.

¹⁴ G. Wentzel, reference 2; result announced in Lecture Series of Norman Bridge Laboratory, Pasadena.

¹⁵ J. Fischer, Ann. d. Physik (5) 8, 821 (1931).

F. Sauter, Ann. d. Physik (5) 9, 217 (1931).

^{&#}x27;7 F. Sauter, Ann. d. Physik (5) 11, 454 (1931), deduces expressions valid when $\beta \sim 1$, $\beta^2 \ll 1$, on the basis of Dirac's relativistic wave-mechanics. The latter expression becomes the same as the one given in this paper when the frequency of the incident x-rays is low.

It is readily seen that both the classical and wave-mechanical curves fit the points as to form but that the asymmetry of the classical curve is unquestionably too weak. We may compare the experimental and theoretical asymmetries by evaluating the quantities defined by the following equations:

$$
\rho = \int_0^{\pi/2} P(\theta) d\theta \Big/ \int_{\pi/2}^{\pi} P(\theta) d\theta;
$$

$$
\theta_0: \int_0^{\theta_0} P(\theta) d\theta = \int_{\theta_0}^{\pi} P(\theta) d\theta;
$$

$$
\overline{\cos \theta} = \int_0^{\pi} P(\theta) \cos \theta d\theta \Big/ \int_0^{\pi} P(\theta) d\theta;
$$

$$
k = \mu \overline{\cos \theta} / (h\nu_0/c), \quad \mu = m\beta c / (1 - \beta^2)^{\frac{1}{2}}.
$$

The last quantity, k , is defined as the ratio of the average forward momentum of the electrons to the momentum of a quantum having an energy equal to that of the electrons. This quantity is particularly useful when the binding energy of the K -electrons is not negligible but is comparable with $h\nu$ of the incident x-rays.

The experimental values of these quantities were obtained by graphical methods from the experimental curve in Fig. 3. These values, with the corresponding theoretical values, are given in Table I. In each case, the experimental value

TABLE I.

	θ o		$\cos \theta$	
Experimental	$81.0^{\circ} + .33^{\circ}$	$1.55 + 0.028$	$0.121 + 0.039$	$1.32 + .042$
Wave mechanics	79.8°	1.72	0.1424	1.57
Auger-Perrin	84.0°	1.37	0.0822	0.90

deviates from the corresponding wave-mechanical value in the direction of weaker asymmetry, while there appears to be real lack of accord between experiment and classical theory.

How does scattering of the electrons affect the experimental values recorded in Table I? We may assume that the nuclear scattering is appreciable over only small angles of scattering. This assumption, combined with the fact that most of the electrons are ejected in the neighborhood of $\theta = 90^{\circ}$, leads to the conclusion that the relative spurious effect of scattering is a maximum in the regions of $\theta = 0^{\circ}$ and $\theta = 180^{\circ}$. This means that the experimental value of $\overline{\cos \theta}$ is affected by scattering, with the change toward a smaller value. Inspection of the experimental curve in Fig. 3 shows, however, that there is a tendency toward compensation, in that scattering raises the curve at both ends. Because of the rough symmetry of the curve about the bipartition angle, $\theta_0 = 81^\circ$, and the fact that, for a given angular range $\delta\theta$, the solid angle is roughly constant in the neighborhood of 81°, it is probable that scattering affects the experimental bipartition angle only inappreciably. A somewhat larger effect is to be expected in the case of ρ , for, while the solid angle for a given range $\delta\theta$ is very nearly constant in the vicinity of $\theta = 90^{\circ}$, the experimental curve is quite asymmetric with respect to this angle and, consequently, the effect of scattering would be to make the value of ρ too small.

The actual differences between the wavemechanical and experimental values of cos θ , θ_0 and ρ bear the ratios to the respective probable errors, 5.7, 3.6 and 6.1. In line with the above argument, θ_0 is in best agreement with theory; but this, of course, does not mean that the whole difference between theory and experiment can be accounted for by scattering.

IV. COMPUTATION OF k from Data Given by OTHER OBSERVERS

It can be shown (with relativity correction) that

$$
k = (1 + 2/\alpha_e)^{\frac{1}{2}} \overline{\cos \theta},
$$

where $\alpha_e = 1/(1 - \beta^2)^{\frac{1}{2}} - 1$. On the basis of wavemechanics,

$$
k = \frac{4}{5} \frac{1}{1+\alpha/2} \cdot \frac{2+\alpha_e}{1+\alpha_e} + \cdots
$$

The factor k is convenient in comparing experiment with theory because its wave-mechanical value is nearly constant over the ordinary range of values of α and α_e . When the binding energy is negligible, $\alpha_e \leq \alpha$ and $k=1.6(1-\beta^2)^{\frac{1}{2}}$, which has the limiting value (when $\beta = 0$), $k = 1.6$. The limiting classical value is 0.8.

Values of k computed from other observers' published values of cos θ are not in satisfactory

Ouantity	Probable error	Values when $N = 400.$ $\gamma = 8/7$		
$\cos \theta_0 \left\{ = \frac{1}{\gamma} \left(1 + \frac{\gamma^2}{2} \right)^{\frac{1}{2}} - 1 \right\}$	$\pm \frac{89.3 \tan \theta_0}{1+2.5\left(\frac{\gamma}{4}\right)^2-5.6\left(\frac{\gamma}{4}\right)^4} \cdot \frac{1}{N^{\frac{1}{2}}}$ percent	0.255 ± 14.5 percent		
θ_0	$\pm \frac{51.2}{1+2.5\left(\frac{\gamma}{4}\right)^2-5.6\left(\frac{\gamma}{4}\right)^4} \cdot \frac{1}{N_{\frac{1}{2}}}$ degrees	$75.2^{\circ}+2.2^{\circ}$		
$\rho = (8+3\gamma)/(8-3\gamma)$	$\pm 67(\rho+1)\left(\frac{1}{\rho N}\right)^{\frac{1}{2}}$ percent	2.5 ± 7.4 percent		
$\overline{\cos \theta} \left[\frac{-\gamma}{k} \right]$	$\pm \frac{67}{\cos \theta (5N)^{\frac{1}{2}}}$ percent	$\begin{cases} 0.229 \pm 6.6 \text{ percent} \\ \pm 6.6 \text{ percent} \end{cases}$		

TABLE II.

accord with wave-mechanical theory. The C. T. R. Wilson method was used in all these investigations and hence, in some cases, the results have been subject to large statistical errors. Assuming the distribution function⁹

$P(\theta) \propto \sin^3 \theta (1 + \gamma \cos \theta),$

(a close approximation to the wave-mechanical distribution when $\gamma = 4\beta/a$, a statistical analysis was carried out for the C. T. R. Wilson method. The results of this analysis are given in Table II in which N is the total number of fog-tracks measured. Application of the analysis to the typical case, $N=400$ and $\gamma = 8/7$ (or $\rho = 2.5$), is included in the tabulation, while in Table III, probable errors in the ordinates of the longitudinal distribution are given. The rather typical range of 15° was used in the calculations for Table III. As Table III is only illustrative of how large the errors are, the calculations have been made for only the forward half of the curve.

TABLE III.

Range (degrees) Percent			$0-15$ 15-30 30-45 45-60		$60 - 75$ 75-90	
(prob. error)	71	22	12	8.3	71	79

Other reasons for the worth of the factor k are that the probable error in this quantity is relatively small, and that, to compute the error, one need know only N and $(\overline{\cos \theta})_{\text{exp}}$. In Table

Ref.	Gas	$\lambda(A)$	β	\boldsymbol{N}	$\cos \theta$	$k_{\rm exp.}$	Probable error	$k_{\rm theor.}$	$k_{\rm theor.}-k_{\rm exp.}$
5	C_2H_5Br	0.586	0.172	233(K)	0.133	1.54	± 0.23	1.56	$+0.02$
3	A in H_2	.709	.231	1200(K)	.169	1.44	\pm .07	1.56	.12 $^{+}$
	A in H_2	.21	.433	1000(K)	.313	1.38	.04 士	1.46	.08 $\mathrm{+}$
3	A in H_2	.21	.433	450(K)	.277	1.22	$\pm .06$	1.46	.24 ┿
	A in H_2	.134	.532			$1.30*$		1.41	.11 \div
	air	.709	.254	272	.182	1.40	±.14	1.55	.15 ┿
	air	.562	.285	200	.210	1.45	± .15	1.54	.09 $^+$
4	N_2	.614	.262	200	.189	1.36	±.15	1.54	.18 $^+$
4	$\rm N_2$.545	.288	93	.179	1.22	\pm .21	1.53	.31 $^{+}$
4	O ₂	.709	.254	159	.202	1.56	\pm .18	1.55	.01 --
4	O ₂	.614	.262	179	.207	1.49	±.16	1.54	.05 \div
4	O ₂	.545	.288	148	.193	1.31	\pm .17	1.53	.22 $+$
Present Work		.7078	.180	22,000(K)	.121	1.32	$\pm .04$	1.57	$+$.25

TABLE IV.

* Value given by Auger for σ , where $\sigma \leq k$ when binding energy is small.

FIG. 4. Comparison of experimental and theoretical values of k . (1) Wave-mechanics, binding energy negligible or small but not negligible; (2) wave-mechanical values multiplied by 0.9; (3) classical theory of Auger-Perrin, binding energy negligible; (4) classical theory appropriate to present work, binding energy not negligible.

IV, experimental values of k , with probable errors computed by the simple formula of Table II, are compared with the appropriate wavemechanical values. These values are plotted in Fig. 4, with twice the probable errors given by the vertical lines.

It is seen that all but one of the experimental values deviate from theory in the direction of weaker asymmetry in the space-distribution. Only one-third of the experimental values include the theoretical values within their respective probable-error ranges, and the probable errors in these particular values are large, varying from 10 percent to 15 percent. The most reliable values (computed from Auger's data) differ from theory by 1.6, 1.9 and 3.9 times the respective probable errors.

The value of k given by the erroneous wavemechanical treatment referred to in the Introduction is just nine-tenths the present wavemechanical value. While this value is of no theoretical significance, it is worthy of note that it is in striking agreement with experiment.

The former theory, regarded as empirical, is represented by the dashed curve (2) in Fig. 4. The average weighted difference between the experimental values (excepting the one point for which the probable error is not known) and the appropriate wave-mechanical values multiplied by 0.9 is $+0.004$ or only 0.3 percent; the corresponding probable error is about 3.0 percent and hence we may say that experiment is in excellent accord with the older, erroneous theory. The deviation from present theory is about 10 percent, or more than three times the probable error, and, consequently, the discrepancy appears real.

An indication that scattering cannot account for the discrepancy is found in the circumstance that no real difference seems to exist between the results of Auger and of Williams, Nuttall and Barlow, in spite of Auger's effort to reduce scattering by diluting the gas studied (argon) in hydrogen, while in the latter work, in which N_2 and O_2 were used, scattering should have been stronger.

In the present work, as we have seen, small angle nuclear scattering affects the bipartition angle less than it does $\overline{\cos \theta}$ or ρ and we may therefore take, as the most reliable asymmetry factor given by the present work, $\theta_0 = 81.0^\circ$ \pm 0.33°. The bipartition angle calculated on the basis of the erroneous theory is 80.8', in very good agreement with the experimental angle.

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