# The Auger Effect in Relative Intensities and Widths of X-Ray Lines

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A systematic study of the widths and relative intensities of certain L lines in the atomic number range  $70 \le Z \le 81$  has been made with a two-crystal spectrometer. It was found that in going from Ta (73) to Tl (81) the width of lines arising from  $L_{\rm I}$  initial states increased in energy by approximately six electron volts, while the widths of lines arising from  $L_{\rm II}$  and  $L_{\rm III}$ initial states increased by only approximately one electron volt. Further, the intensities of  $L_{\rm I}$  lines relative to the intensities of  $L_{\rm II}$  and  $L_{\rm III}$  lines showed a marked decrease with atomic number. Both of these variations can be attributed to the rapid increase with atomic number of the probabilities of the Auger transitions  $L_{\rm I} \rightarrow L_{\rm III} M_{\rm IV}$  and  $L_{\rm I} \rightarrow L_{\rm III} M_{\rm V}$ . The results on line widths are in accord with the theory of Weisskopf and Wigner. All the data recorded indicate that Auger transitions are of prime importance in determining the relative intensities and widths of certain x-ray lines.

# I. INTRODUCTION

THAT the Auger effect plays an important part in determining both the relative intensities and the widths of x-ray lines was first recognized by Coster and Kronig.<sup>1</sup> Auger transitions, otherwise known as radiationless transitions, increase the widths of lines since they decrease the lifetime of the states; secondly, they decrease the relative intensities of diagram lines since Auger transitions allow an atom to leave a state without emitting a quantum; thirdly, they increase the relative intensities of "second-order" lines, otherwise known as "satellite" lines, since the final states of many Auger transitions are the initial states for satellite emission.

The importance of Auger transitions in x-ray spectroscopy is increased by the fact that their probabilities often vary rapidly from one element to the next. Accordingly, the Auger effect provides exactly the sort of mechanism required to explain the puzzling variations that have been observed in line widths and in relative intensities. Indeed, this rapid variation of Auger probabilities with atomic number makes it possible to distinguish the results of Auger transitions from the effects of other processes.

The present paper reports systematic measurements of the widths and relative intensities of certain *L*-series lines for elements of atomic number between 73 and 81. In this region it is known that the probabilities of the Auger transitions from an  $L_{\rm I}$  initial state to the final states of double ionization  $L_{\rm III}M_{\rm IV}$  and  $L_{\rm III}M_{\rm V}$  are rapidly changing with atomic number. Therefore, one may expect appreciable changes in relative intensities and widths of lines arising from  $L_{\rm I}$ transitions. The expected variations are clearly shown by the data reported here.

### II. THEORY OF SHAPES AND WIDTHS OF X-RAY LINES

On the assumptions of the Dirac radiation theory Weisskopf and Wigner<sup>2</sup> found that the frequency distribution of a line arising from transitions between the states A and B was given by

$$J(\nu)\Delta\nu = \frac{(\Gamma^A + \Gamma^B)\Delta\nu}{[(\Gamma^A + \Gamma^B)/2]^2 + 4\pi^2(\nu - \nu_B{}^A)^2}$$

Here  $\Gamma^{A}$  and  $\Gamma^{B}$  are, respectively, the reciprocals of the mean life<sup>3</sup> of atoms in states A and B, and  $\nu_{B}{}^{A} = (E_{A} - E_{B})/\hbar$  where  $E_{A}$  and  $E_{B}$  are the most probable energies of states A and B. The full width at half maximum of this distribution is  $\Delta \nu = (\Gamma^{A} + \Gamma^{B})/2\pi$  in frequency units, or  $\Delta E = \hbar (\Gamma^{A} + \Gamma^{B})$  in ergs.

The width of a line is seen to be proportional to the sum of the reciprocals of the mean lifetimes

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<sup>&</sup>lt;sup>1</sup> Coster and Kronig, Physica 2, 14 (1935).

<sup>&</sup>lt;sup>2</sup> Weisskopf and Wigner, Zeits. f. Physik **63**, 54 (1930). <sup>3</sup> The mean life of atoms in the state A is here defined

as the time required for N atoms in that state at time t=0 to be reduced to 1/e of N (e is the Naperian base).

of the initial and final states. It is convenient to interpret  $\hbar\Gamma^{A}$  and  $\hbar\Gamma^{B}$  as the energy widths of states A and B, respectively. Speaking in these terms, one may say that the width of a line is equal to the sum of the widths of the initial and final states.

The reciprocal of the mean life of a state  $\Gamma^{A}$  is equal to the sum of the probabilities of all spontaneous transitions from state A to any other state, so one may write

$$\Gamma^{A} = \sum_{B} \gamma_{B}^{A},$$

where  $\gamma_{B^{A}}$  is the probability for the transition  $A \rightarrow B$ , and the sum extends over all possible transitions. Taking account of both radiative and Auger processes, one may rewrite the above in the form

$$\Gamma^{A} = \sum_{R} \gamma_{R}^{A} + \sum_{S} \gamma_{S}^{A},$$

where  $\gamma_{R^{A}}$  is the transition probability for the radiative transition  $A \rightarrow R$  and  $\gamma_s^A$  is the probability for the Auger transition  $A \rightarrow S$ .

Several investigators have made calculations on the relative importance of radiative and Auger transitions in determining the widths of energy levels. Ramberg and Richtmyer<sup>4</sup> have computed both  $\sum_{R} \gamma_{R}^{A}$  and  $\sum_{S} \gamma_{S}^{A}$  for the K,  $L_{I}$ ,  $M_{I}$ ,  $M_{II}$ ,  $M_{\rm III}$ , and  $N_{\rm I}$  levels for Au (79). They found that the radiative process accounts for almost all the transitions from the K shell, but that for all other levels investigated, the Auger contribution to the level width was dominant. For the  $L_{I}$  level of gold they found the ratio of Auger transition probabilities to radiative transition probabilities to be six to one. Pincherle,<sup>5</sup> working on gold but using hydrogenic wave functions, also found this ratio to be roughly six to one, although his calculated value of the width was less than half that of Ramberg and Richtmyer. Such data as are available on fluorescence yields confirm the idea that Auger transitions are often far more probable than radiative ones.

Further, an Auger transition may be highly probable for one element, and forbidden for an element adjacent in the periodic table.6 Any

abrupt change in the probability of a given Auger transition should result in a corresponding change in the width of the initial state and hence in a variation of the widths of all x-ray lines associated with that state. Thus, for a series of elements, one may study the changes in the probability of an Auger transition through observations of the variation in the widths of selected x-ray lines.

## III. THE AUGER TRANSITIONS $L_{I} \rightarrow L_{III}M_{IV,V}$

The two Auger transitions that are the primary concern of this paper have a common  $L_{I}$  initial state and their final states (of double ionization) are  $L_{III}M_{IV}$  and  $L_{III}M_{V}$ . These radiationless transitions are possible whenever the energy of the  $L_{I}$  state is greater than the energies of the states  $L_{III}M_{IV}$  and  $L_{III}M_{V}$ , respectively. In Fig. 1 the difference in the  $\nu/R$  values of the  $L_{\rm I}$  and  $L_{\rm III}$ states is plotted as a function of atomic number; on the same graph, the  $\nu/R$  values for the  $M_{\rm IV}$ and  $M_{\mathbf{v}}$  states of element Z+1 are also plotted. It is clear from the figure that the Auger transition  $L_{I} \rightarrow L_{III} M_{V}$  can take place for elements of atomic number below 50 and above 73, while the transition  $L_{I} \rightarrow L_{III} M_{IV}$  is confined to elements of atomic numbers below 50 or above 77.

If the probabilities  $\gamma_{L_{111}M_{1V}}^{L_1}$  and  $\gamma_{L_{111}M_{V}}^{L_1}$  become sufficiently large, they may constitute the principal factor in fixing the width of the  $L_{I}$  level. Calculations of Ramberg and Richtmyer<sup>4</sup> indicate that these two transitions alone contribute 62 percent of the width of the  $L_{\rm I}$  level for gold. For elements of atomic number between 50 and 72 these transitions are forbidden and so contribute inappreciably. On the basis of these considerations one should expect a large change in the width of the  $L_{I}$  level for elements between 72 and 79.

Data on the satellites of the  $L\alpha$  doublet confirm the importance and high probabilities of the Auger transitions  $L_{I} \rightarrow L_{III} M_{IV,V}$ . The  $L\alpha$  satellites, according to Druyvestyn<sup>7</sup> and others, arise from transitions from the states of double ioniza-

<sup>&</sup>lt;sup>4</sup> Ramberg and Richtmyer, Phys. Rev. 51, 913 (1937). <sup>5</sup> L. Pincherle, Nuovo Cimento (N.S.) 81, 162 (1935).

<sup>&</sup>lt;sup>6</sup> This is true because such a transition is possible only when the energy of the initial state is greater than that of

the final state (of double ionization) and is most probable when this energy difference is not too great. When the energy difference is very large, the probability of the Auger transition is small, since the wave function describing the ejected electron oscillates too rapidly to make the transition probability integral large.

<sup>&</sup>lt;sup>7</sup> M. J. Druyvestyn, Zeits. f. Physik 43, 707 (1927).



FIG. 1. Coster-Kronig diagram. The Auger transition  $L_{I} \rightarrow L_{III} M_{IV}$   $(L_{I} \rightarrow L_{III} M_{V})$  is possible for an element of atomic number Z if the energy difference  $L_{I} - L_{III}$  is greater than the energy of the  $M_{IV}(M_V)$  level of element Z+1.

tion  $L_{III}M_{IV}$  and  $L_{III}M_{V}$ . Early observations of these satellites showed that they were intense relative to the  $L\alpha$  doublet for elements of atomic number above 73 and below 50, while they were very weak for elements in the atomic number range  $50 \le Z \le 73$ . The explanation of this intensity anomaly was proposed by Coster and Kronig<sup>1</sup> who suggested that the high probability of the Auger transitions  $L_{I} \rightarrow L_{III} M_{IV,V}$  for elements below 50 and above 73 should result in a great increase in the number of atoms which reach the initial state for  $L\alpha$  satellite production. Hirsh<sup>8</sup> measured photographically the relative intensities of the  $L\alpha$  satellites in the atomic number range  $40 \le Z \le 52$ . His data confirmed the prediction of Coster and Kronig, as have subsequent measurements on the  $L\alpha$  satellites by Randall and Parratt,9 who used a two-crystal spectrometer.

Shrader,<sup>10</sup> also using a two-crystal spectrometer, has determined the relative intensities of the  $L\alpha$  satellites for elements of atomic number between 73 and 92. His results are shown in

Fig. 2. The curve fitting his data is arbitrarily resolved into two components, one assigned to each of the transitions  $L_{I} \rightarrow L_{III} M_{V}$  and  $L_{I} \rightarrow L_{III} M_{IV}$  which are possible for elements of atomic number greater than 72 and 76, respectively.

If one assumes that the sum of the probabilities of the Auger transitions  $L_{I} \rightarrow L_{III} M_{IV,V}$  is roughly proportional to the relative intensity of the  $L\alpha$  satellites, it is apparent that the sum  $\gamma_{{\cal L}_{III}{\cal M}_{IV}}^{{\cal L}_{I}} + \gamma_{{\cal L}_{III}{\cal M}_{V}}^{{\cal L}_{I}}$  should increase rapidly with atomic number for  $73 \le Z \le 81$ . As a consequence, the width of the  $L_{I}$  level should increase with atomic number in this range. Data presented in the present paper confirm this assumption and prediction.

Any increase in the probabilities of Auger transitions from the  $L_{\rm I}$  state should result in a decrease in the intensities of  $L_{I}$  lines relative to the intensities of  $L_{II}$  and  $L_{III}$  lines, provided (1) there exists no corresponding abnormal behavior in Auger transitions from the  $L_{II}$  and  $L_{III}$ states, and (2) the ratio of the numbers of atoms reaching the  $L_{I}$ ,  $L_{II}$ , and  $L_{III}$  states is constant. This prediction is based on the obvious fact that the relative number of atoms leaving the  $L_{I}$  level by radiative transitions must decrease as

<sup>&</sup>lt;sup>8</sup> F. R. Hirsh, Phys. Rev. 48, 722 (1935).
<sup>9</sup> Randall and Parratt, Phys. Rev. 57, 786 (1940).
<sup>10</sup> R. E. Shrader, unpublished work. The author is grateful to Dr. Shrader for the use of his unpublished curve.

the Auger probabilities increase. Coster and De Langen<sup>11</sup> have shown that the intensities of the  $L_{\rm I}$  lines,  $L\beta_3$  and  $L\beta_4$ , are smaller relative to the intensity of the  $L_{\rm II}$  line  $L\beta_1$  for Ag (47) than for Sn (50) and Sb (51). The probabilities of the Auger transitions  $L_{\rm I} \rightarrow L_{\rm III} M_{\rm IV,V}$  are much greater for silver than for tin and antimony, so their data are in agreement with expectations. Data presented in this paper reveal a marked decrease with atomic number in the intensity of the  $L_{\rm I}$  lines relative to the  $L_{\rm II}$  and  $L_{\rm III}$  lines from Ta (73) to Tl (81).

### **IV. EXPERIMENTAL**

The direct-reading two-crystal spectrometer used in the present work has been described by Richtmyer and Barnes.<sup>12</sup>

The ionization chamber, 10 inches long, was filled with argon to a pressure of three atmospheres. At one angstrom the chamber absorbed 99 percent of the incident intensity. Special provision was made to collect ions produced near the entrance window before re-combination. The ionization currents were amplified in a Brown-DuBridge circuit, with an FP-54 electrometer tube mounted adjacent to the ionization chamber. Either of two input resistors, of the order of 10<sup>10</sup> and 10<sup>11</sup> ohms, respectively, could be placed in the control-grid circuit.

The x-ray tube was of the Coolidge type with demountable filament and target assemblies. Because the target was not shielded from the filament, the maximum ordinate was checked after recording each spectral curve to make certain that no appreciable tungsten layer had been deposited on the target. The focal spot was roughly  $10 \times 5$  mm. The x-rays made an angle of thirty degrees with the target face; the cathode electrons, sixty degrees. The tube window was a disk of beryllium which transmitted 84 percent of the incident intensity at 1.274A (Au  $L\alpha_1$ ).

The targets used were prepared as follows: Yt (70) and Lu (71) powdered samples, reported to be "alloyed" with aluminum, were pressed into a thin layer of lead (82) which had been melted onto copper and silver, respectively. There were definite x-ray spectral indications

that other rare earths were also present in the samples. These targets were not entirely satisfactory and the power dissipation had to be limited to approximately 300 watts in each case. The Ta (73), W (74), and Pt (78) targets were made by hard soldering a strip of each to copper. Crushed metallic bits of Rh (75), Os (76), and Ir (77) were forced into silver with a hydraulic press. A thin foil of Au (79) was hard-soldered to 25-ml silver. These targets were entirely satisfactory for powers up to one kilowatt. The Th (81) target was made by evaporating thallium in vacuum and allowing it to condense on a sheet of 10-ml copper which had been previously soft soldered to the target carriage. The evaporation was carried out in a tube designed to hold the entire target carriage, so that it would not be necessary to heat the thallium once it had been deposited. It was necessary to limit the x-ray tube power to 500 watts to prevent the reevaporation (or sputtering) of the thallium.

The data for all lines of a given element were recorded at the same x-ray tube voltage. For every element except thallium, this voltage was approximately three times the excitation potential of the  $L_{\rm I}$  state. With the thallium target the tube was often unstable at this potential (46 kv) so all thallium data were taken at 42 kv. Tube currents varied from one to thirty milliamperes. Both the tube voltage and current were maintained con-



FIG. 2. The intensity of the  $L\alpha$  satellites relative to the  $L\alpha$  doublet as a function of atomic number. The curve *abe* represents the contribution of the Auger transition  $L_{I} \rightarrow L_{III} M_{V}$ , while the curve *fgh* is attributed to the transition  $L_{I} \rightarrow L_{III} M_{IV}$ . (Data taken by Dr. R. E. Shrader.)

<sup>&</sup>lt;sup>11</sup> Coster and De Langen, Physica 3, 282 (1936).

<sup>&</sup>lt;sup>12</sup> Richtmyer and Barnes, Rev. Sci. Inst. 5, 351 (1934).

stant within 1.5 percent by manual control throughout the recording of each spectral curve.

### V. CRYSTALS

The observed widths of x-ray lines should be corrected for the finite resolving power of the spectrometer. For a two-crystal spectrometer the resolving power depends on (1) the geometrical divergence of the x-ray beam and (2) the diffraction patterns of the crystals. Parratt<sup>13</sup> and others have shown that with slits properly arranged the resolving power of the spectrometer is limited by the crystal diffraction patterns. For the data presented here the geometrical resolving power was 250,000. This is approximately 25 times as great as the physical resolving power of the instrument, as determined by the diffraction patterns of the crystals.

The correction to the observed width of a line may be a function of the width and shape of the line in question, as well as of the diffraction patterns of the crystals. Several correction formulae have been proposed and used for correcting widths reported in the literature, but as a general formula each is open to question.

On the approximations (a) that the diffraction patterns of the crystals in both the (1, +1) and (1, -1) positions are identical, and (b) that their shapes and the true x-ray line shape may each be represented by a Gaussian error function, it has been shown that

$$W_T^2 = W_0^2 - W_c^2,$$

where  $W_T$  is the true width of the line, and  $W_0$ 

TABLE I. Data on condition of calcite crystals.

Date	Observer	Radiation	(1, +1) full width x.u.	(1, -1) full width x.u.	Per- cent reflec- tion
Dec. 1938 Apr. 1939 Oct. 1939 June 1940 June 1940	Shrader Parratt Cooper* Cooper Cooper	CuKα 1.54A MoKα 0.71A MoKα 0.71A MoKα 0.71A CuKα 1.54A	0.38 0.36 0.365 0.57	0.15 0.087 0.088 0.090 0.152	61 48.5 56.5 55 60.5

<sup>\*</sup> Between the measurements of Parratt and those of Cooper the crystals were ground and etched several times, following the method of Manning [Rev. Sci. Inst. 5, 316 (1934)]. The increase in the widths of the (1, +1) and (1, -1) curves and the decrease of the percent reflection from October, 1939 to June, 1940 is probably due to general deterioration of the crystal faces, although the differences are all of the order of the uncertainties involved in the measurements.

and  $W_c$  are the observed widths in the (1, +1)and (1, -1) positions, respectively. Since  $W_0$  for *L*-series lines is considerably greater than  $W_c$ , one may write

$$W_T = W_0 [1 - (W_c/W_0)^2]^{\frac{1}{2}},$$
  
$$\cong W_0 - (W_c^2/2W_0).$$

Thus for Gaussian error shapes the correction is proportional to the square of the (1, -1) width and inversely proportional to the width of the observed (1, +1) curve.

If one assumes the shapes to be given by the classical dispersion function, the correction formula may be written in the simple form :

$$W_T = W_0 - W_c.$$

Here the correction is simply the width of the (1, -1) curve and is independent of the observed width of the line. The correction for the classical dispersion shapes is always greater than that for the Gaussian error shapes.

There is much experimental evidence that the (1, -1) shapes of most lines in the wave-length region  $0.5A \le \lambda \le 3.0A$  lie somewhere between the classical dispersion and the Gaussian error shapes. Parratt and Miller<sup>14</sup> have shown that the square of the classical dispersion curve gives a reasonably good fit for a large number of observed (1, -1) shapes in this wave-length region. The fact that the shapes of observed (1, -1) curves lay between the Gaussian error and classical dispersion shapes led Richtmyer, Barnes, and Ramberg<sup>15</sup> to propose the correction formula

$$W_T = W_0 - \frac{1}{2}W_c,$$

which gave more consistent values for  $W_T$  of certain lines than did either of the formulae above.

In the search for a better established correction formula Parratt<sup>16</sup> made extensive measurements on the widths of certain  $K\alpha$  doublets in various parallel and anti-parallel positions for 16 pairs of crystals. He found that for several pairs of crystals one could write  $W_T = W_0 - 29W_c^{1.7}$  where  $W_T$  is the true width,  $W_c$  the (n, -n) width, and

<sup>&</sup>lt;sup>13</sup> L. G. Parratt, Phys. Rev. 46, 749 (1934).

<sup>&</sup>lt;sup>14</sup> Parratt and Miller, Phys. Rev. 49, 280 (1935).

<sup>&</sup>lt;sup>15</sup> Richtmyer, Barnes, and Ramberg, Phys. Rev. **46**, 843 (1934).

<sup>&</sup>lt;sup>16</sup> L. G. Parratt, Rev. Sci. Inst. 6, 387 (1935).

#### AUGER EFFECT

TABLE II. Widths of *L*-series lines.

Line	Transition	λ (x.u.)	Δλ obs. (x.u.)	Δλ cor. (x.u.)	ΔV cor. ev	Estimated Error in Δλ obs.	Line	Transition	λ (x.u.)	Δλ obs. (x.u.)	Δλ cor. (x.u.)	$\Delta V$ cor. ev	Estimated Error in Δλ obs.
Ytter $L\beta_3$ $L\beta_4$ $L\gamma_3$ $L\beta_1$	bium (70) $L_{I}M_{III}$ $M_{II}$ $N_{III}$ $L_{II}M_{IV}$	1449.4 1488.2 1219.8 1472.5	2.32 2.63 1.34 1.41	2.21 2.51 1.23 1.30	12.9 13.9 10.2 7.4	0.08 0.08 0.05 0.03	$\begin{vmatrix} \overline{\text{Osmin}} \\ L\gamma_3 \\ L\beta_1 \\ L\gamma_1 \\ L\gamma_6 \\ L\gamma_6 \end{vmatrix}$	$ \begin{array}{c} \text{um (76).} \\ \hline M \text{III} \\ L_{\text{II}} M_{\text{IV}} \\ N_{\text{IV}} \\ \hline N_{\text{IV}} \\ O_{\text{IV}} \\ I \\ \end{array} $	Continued 989.8 1194.5 1022.8 998.8	1.11 0.88 0.91 0.76	1.00 0.77 0.80 0.65	12.6 6.6 9.4 6.5	0.03 0.02 0.02 0.05
Luteo	cium (71)						$L\beta_2$	$L_{III}N_{V}$	1100.9	1.21	1.10	9.9	0.03
$Leta_3\ Leta_1\ Leta_2$	$\begin{array}{c} L_{\rm I}M_{\rm III}\\ L_{\rm II}M_{\rm IV}\\ L_{\rm III}N_{\rm V}\end{array}$	1398.2 1420.7 1367.2	2.15 1.21 1.65	$2.04 \\ 1.10 \\ 1.54$	12.8 6.7 10.1	0.15 0.03 0.05	$ \begin{array}{c c} \text{Iridiu} \\ L\beta_3 \\ L\beta_4 \\ L\gamma_2 \end{array} $	m (77) $L_I M_{III}$ $M_{II}$ $N_{II}$	1138.5 1177.2 963 3	1.68 2.10 1.20	1.57 1.99 1.10	14.9 17.7 14 6	$0.04 \\ 0.06 \\ 0.04$
Tant: $L\beta_3$ $L\beta_4$ $L\gamma_2$ $L\gamma_3$ $L\beta_4$	alum (73) <i>L</i> <sub>I</sub> <i>M</i> <sub>III</sub> <i>M</i> <sub>II</sub> <i>N</i> <sub>III</sub> <i>N</i> <sub>III</sub> <i>L</i> = <i>M</i> ==	1304.1 1343.1 1103.0 1097.1	1.73 2.18 1.26 1.09	1.62 2.07 1.15 0.98	11.7 14.1 11.6 10.0	0.03 0.04 0.03 0.03	$ \begin{array}{c} L\gamma_{2} \\ L\gamma_{3} \\ L\beta_{1} \\ L\gamma_{1} \\ L\gamma_{6} \\ L\beta_{2} \end{array} $	$N_{III}$ $L_{II}M_{IV}$ $N_{IV}$ $O_{IV}$ $L_{III}N_{V}$	957.1 1155.4 988.8 964.9 1133.0	1.09 0.85 0.87 0.70 1.12	0.99 0.74 0.76 0.59 1.01	13.3 6.8 9.6 7.8 9.7	0.03 0.02 0.02 0.05 0.03
$L \beta_1 \\ L \gamma_1 \\ L \gamma_6 \\ L \beta_2$	$L_{III}M_{IV}$ $N_{IV}$ $O_{IV}$ $L_{III}N_{V}$ $(74)$	1324.2 1135.6 1111.5 1281.9	$   \begin{array}{r}     1.03 \\     1.11 \\     0.70 \\     1.42   \end{array} $	1.00 0.59 1.31	9.5 7.3 9.8	0.02 0.03 0.04 0.03	Platin $L\beta_3$ $L\beta_4$ $L\gamma_3$	$L_{I}M_{III}$ $M_{III}$ $M_{III}$ $N_{III}$	1101.7 1139.9 926.0	1.70 2.05 1.09	1.59 1.94 0.98	16.1 18.4 14.1	0.17 0.09 0.03
$L\beta_3$ $L\beta_4$ $L\gamma_2$ $L\gamma_3$	$\begin{array}{c} \text{sten} (74) \\ L_I M_{III} \\ M_{II} \\ N_{III} \\ N_{III} \end{array}$	1259.9 1298.8 1065.9 1059.9	1.70 2.18 1.23 1.11	1.59 2.07 1.12 1.00	12.3 15.1 12.1 11.0	0.03 0.04 0.03 0.03	$\begin{vmatrix} L\beta_1 \\ L\gamma_1 \\ L\beta_2 \end{vmatrix}$	$L_{\mathrm{II}} M_{\mathrm{IV}}^{\mathrm{IIV}}$ $N_{\mathrm{IV}}$ $L_{\mathrm{III}} N_{\mathrm{V}}$	1117.6 956.0 1099.7	$0.84 \\ 0.85 \\ 1.10$	0.73 0.75 0.99	7.2 10.1 10.1	0.02 0.02 0.04
$Leta_1 \ L\gamma_1 \ L\gamma_6 \ Leta_2$	$\begin{array}{c} L_{\rm II} M_{\rm IV} \\ N_{\rm IV} \\ O_{\rm IV} \\ L_{\rm III} N_{\rm V} \end{array}$	1279.2 1096.3 1072.1 1242.0	0.97 1.02 0.64 1.34	0.86 0.91 0.53 1.23	6.5 9.3 5.7 9.8	0.02 0.02 0.04 0.03	$\begin{vmatrix} \text{Gold} \\ L\beta_3 \\ L\beta_4 \\ L\gamma_2 \\ L\gamma_3 \end{vmatrix}$	$(79) \\ L_{I}M_{III} \\ M_{II} \\ N_{III} \\ N_{V-V}$	1065.5 1104.2 902.5 805.8	$1.70 \\ 2.05 \\ 1.20 \\ 1.07$	1.59 1.94 1.10	17.2 19.6 16.6	0.15 0.09 0.05 0.03
Rhen	ium (75)						$L\gamma_3$ $L\beta_1$	$L_{\rm II} M_{\rm IV}$	1081.3	0.81	0.70	7.4	0.03
$Leta_3 \ Leta_4 \ L\gamma_2 \ L\gamma_3$	$L_{I}M_{III}$ $M_{II}$ $N_{II}$	1217.6 1256.3 1029.9	1.75 2.22 1.25	1.64 2.11 1.14	13.6 16.4 13.2	0.03 0.04 0.03	$egin{array}{c} L\gamma_1 \ L\gamma_6 \ L\beta_2 \end{array}$	$N_{IV} O_{IV} D_{IV} L_{III} N_{V}$	924.6 901.0 1068.0	0.80 0.60 1.04	0.70 0.50 0.93	10.1 7.6 10.1	0.02 0.04 0.04
$L\gamma_3$ $L\beta_1$ $L\gamma_1$ $L\gamma_6$ $L\beta_2$	$L_{II} M_{IV} M_{IV} N_{IV} O_{IV} L_{III} N_{V}$	1023.0 1236.0 1058.7 1034.4 1204.1	0.92 0.98 0.69 1.27	0.81 0.87 0.58 1.16	6.5 9.5 6.7 9.8	0.03 0.02 0.02 0.05 0.03	$\begin{vmatrix} \text{Thall} \\ L\beta_3 \\ L\beta_4 \\ L\gamma_2 \end{vmatrix}$	$\begin{array}{c} { m ium} \ (81) \ L_{\rm I} M_{ m III} \ M_{ m II} \ N_{ m II} \end{array}$	998.5 1037.0 845.7	1.60 1.89 1.12	1.49 1.78 1.02	18.4 20.4 17.5	0.06 0.05 0.04
Osmi $L\beta_3$	um (76) $L_{I}M_{III}$ $M_{II}$	1176.8	1.77	1.66	14.7	0.03	$ \begin{array}{c c} L\gamma_3 \\ L\beta_1 \\ L\gamma_1 \\ L\gamma_1 \end{array} $	$\begin{array}{c} N_{\rm III} \\ L_{\rm II} M_{\rm IV} \\ N_{\rm IV} \\ O_{\rm TV} \end{array}$	839.3 1013.0 865.7 842.3	1.02 0.75 0.71	0.92 0.64 0.61	16.1 7.7 10.0	0.04 0.02 0.02
$\widetilde{L}_{\gamma_2}$	NII	995.9	1.24	1.13	14.0	0.04	$\begin{vmatrix} L & \gamma & 6 \\ L & \beta_2 \end{vmatrix}$	$L_{\rm III} \overset{OIV}{N_{\rm V}}$	1008.2	0.95	0.84	10.2	0.04

 $W_0$  the (n, +n) width (all widths in x.u.). Crystals satisfying this relation are said to be of Class I; those which do not are grouped in class II.

Data on the crystals in the present work (3A and 3B, used by Richtmyer, Barnes, and Ramberg,<sup>15</sup> and by Shrader<sup>10</sup>) are recorded in Table I. Comparing these data with those of Parratt, one sees that the crystals are rather poor at Mo  $K\alpha$  (0.71A) and good at Cu  $K\alpha$  (1.54A). The crystals do not fall into class I. Consequently the correction formula proposed by Parratt for crystals of class I is not applicable and one must resort to some other scheme for correcting the

observed widths. The corrections applied to the observed widths reported here were determined as follows: Parratt's values of the true widths of the Mo  $K\alpha$  and the Cu  $K\alpha$  lines were accepted as standards. Then the corrections for Mo  $K\alpha$  was 0.10 x.u. and for Cu  $K\alpha$  0.12 x.u. It was then assumed (a) that the correction was a linear function of the wave-length in the range from 0.71A to 1.54A and (b) that the correction is a function of wave-length alone. The first assumption cannot introduce appreciable error, but the second is open to question. It was made only because it is not yet known how the correction should be influenced by the width.





FIG. 3. Widths of *L*-series lines in electron volts as a function of atomic number. The lines  $L\beta_3$ ,  $L\beta_4$ ,  $L\gamma_2$ , and  $L\gamma_3$  arise from  $L_{\rm I}$  initial states;  $L\beta_1$  and  $L\gamma_1$  are  $L_{\rm II}$  lines, and  $L\beta_2$  has an  $L_{\rm III}$  initial state.

### VI. WIDTH DATA

Data on the width of certain L lines of ten elements are presented in Table II. The observed widths in x.u. were those measured after correction for zero drift of the amplifying circuit, for the background of continuous radiations, and for overlapping of neighboring lines. The "corrected" widths were obtained by subtracting a correction for the diffraction patterns of the crystals. In the last column of Table II are the estimated errors in the observed widths. Lines with large errors were either very weak or badly overlapped by neighboring lines.

In Fig. 3 the corrected widths, in electron volts, of seven L lines are plotted as a function of atomic number. Of these seven lines,  $L\beta_3$ ,  $L\beta_4$ ,  $L\gamma_2$ , and  $L\gamma_3$  arise from transitions from an  $L_{\rm I}$  initial state, while  $L\beta_1$  and  $L\gamma_1$  have an  $L_{\rm II}$  initial state, and  $L\beta_2$  arises from a transition from the  $L_{\rm III}$  state. For elements 73 to 81, the lines arising from the  $L_{\rm I}$  initial state show increases in widths of approximately six electron volts while the lines arising from  $L_{\rm II}$  and  $L_{\rm III}$  initial states have changes in width of the order of one electron volt. Thus, as predicted by consideration of the Auger effect, the  $L_{\rm I}$  lines show rapid increases

in widths for elements 73 to 81, while  $L_{II}$  and  $L_{III}$  lines show but small changes.

Since each of the  $L_{I}$  lines has so nearly the same increment in width, it seems conclusive that an

TABLE III. Comparison of present width data with that of other observers.

Element	Line	Cooper	Williams*	R. B. R. <sup>14</sup>
W(74)	$L\beta_1$	0.97	0.94	
. ,	$L\beta_2$	1.34	1.38	
	$L\gamma_1$	1.02	1.01	
Ir(77)	$L\beta_1$	0.85	0.82	
. ,	$L\beta_{2}$	1.12	1.17	
	$L\gamma_1$	0.87	0.92	
Pt(78)	$L\beta_1$	0.84	0.80	
	$L\beta_2$	1.10	1.14	
	$L_{\gamma_1}$	0.85	0.84	
Au(79)	$L\beta_1$	0.81	0.79	0.81
(,	$L\beta_2$	1.04	1.06	1.04
	$L\beta_3$	1.70		2.00
	$L\beta_4$	2.05		2.00
	$L_{\gamma_1}$	0.80	0.79	0.82
	$L\gamma_2$	1.20		1.20
	$L\gamma_3$	1.07		1.06
	$L\gamma_6$	0.60		0.60
Tl(81)	$L\beta_1$	0.75	0.74	
	$L\beta_2$	0.95	1.00	
	$L_{\gamma_1}$	0.71	0.74	

\* J. H. Williams, Phys. Rev. 45, 71 (1934).

increase in the width of the  $L_{\rm I}$  state is responsible for the major part of this variation. It is true that some small portion of the width changes may be attributed to changes in the breadths of the final states, and indeed, several other factors may have some influence on the line widths. Nevertheless, the conclusion stands that the observed variations are due primarily to the change in the width of the  $L_{\rm I}$  level and that all other factors are of secondary importance.

A few of the lines measured by the author have also been measured by other observers. A comparison of the widths reported in this paper with those of the other observers is given in Table III. The observations agree within the experimental error for every line except Au  $L\beta_3$ . This line overlaps Au  $L\beta_2$  so badly that an arbitrary graphical resolution must be carried out. Richtmyer, Barnes, and Ramberg<sup>15</sup> used only classical dispersion curves for  $L\beta_2$  and  $L\beta_3$  and neglected the "residuals," whereas the author adjusted the shapes of the two lines to fit the observed contour.

### VII. RELATIVE INTENSITIES

The relative intensities are assumed to be proportional to the product of the energy width and the relative maximum ordinate of each spectral curve. Since all the lines compared had approximately the same (classical dispersion) shape, and nearly the same wave-length, the uncertainty introduced by this approximation is believed to be less than that involved in the comparison of maximum ordinates. None of the numerous and troublesome corrections required for the most precise comparisons of relative intensities has been made.

TABLE IV. Relative intensities of L-series lines.

	1			1		
		Relative t	to $L\beta_1$	R	elative to	L
	Lβ₂	L <sub>β</sub>	Lβ4	$L\gamma_2$	$L\gamma_1 = 100$ $L\gamma_3$	΄ Lγ6
Yt(70)		(14.8-?)	(13.0-?)			
Lu(71)	44.4	(15.7 - ?)	· ·			
Ta(73)	46.1	18.5	13.4	17.3	25.1	2.0
W(74)	46.2	16.2	13.8	14.1	20.4	2.8
Rh(75)	44.5	15.2	11.4	13.3	18.5	4.7
Os(76)	45.8	13.9	11.1	11.5	17.0	6.2
Ir(77)	45.7	14.2	10.7	12.4	15.5	7.3
Pt(78)	43.7	14.1	9.9			
Au (79)	45.0	13.0	10.1	10.5	14.1	11.3
Tl(81)	45.1	9.4	8.8	11.0	13.1	12.3
				l		

Two methods for comparing maximum ordinates were used. First, the spectrometer was set on the peak of each line in succession with the power in the x-ray tube and the sensitivity of the recorder kept constant. After subtracting the background of continuous radiation and the contribution of overlapping lines, the relative maximum ordinates were measured. Second, the maximum deflection produced by each of the lines was made approximately full scale and the



FIG. 4. The intensities of  $L\beta_3$  and  $L\beta_4$  relative to  $L\beta_2$ .  $L\beta_3$  and  $L\beta_4$  arise from  $L_{\rm I}$  initial states while  $L\beta_2$  has the initial state  $L_{\rm III}$ .

relative maximum ordinate for each was computed by the use of the known ratios of amplifier sensitivities and of the measured x-ray tube currents. An average of the relative maximum ordinate given by these two methods was used in computing the relative intensities.

In Table IV the areas (products of maximum ordinate and width) of  $L\beta_2$ ,  $L\beta_3$  and  $L\beta_4$  relative to the area of  $L\beta_1$  ( $L\beta_1=100$ ) are recorded. The greatest wave-length separation of any of these lines from  $L\beta_1$  is 53.5 x.u. It is estimated that the values reported are accurate to within two percent of the area of  $L\beta_1$ . In Fig. 4 the intensities of  $L\beta_3$  and  $L\beta_4$  are plotted relative to  $L\beta_2$  which is an  $L_{III}$  line. These data show that the intensities of the two  $L_I$  lines are decreasing relative both to the  $L_{II}$  line and to the  $L_{III}$  line in the atomic number range  $73 \le Z \le 81$ .

The intensities of  $L\gamma_2$ ,  $L\gamma_3$ , and  $L\gamma_6$  relative to  $L\gamma_1$  are also recorded in Table IV. The maximum wave-length separation from  $L\gamma_1$  is 38.5 x.u. and the estimated error is approximately two percent of the intensity of  $L\gamma_1$ . [The line  $L\gamma_6$  ( $L_{\rm II}O_{\rm IV}$ ) is not observed for elements of atomic number below 70, but rapidly increases in relative intensity from Ta (73) to Tl (81). Its behavior is quite extraneous to the topic being discussed.]

Once again, it is clear that the lines arising from  $L_{I}$ transitions  $(L\gamma_2 \text{ and } L\gamma_3)$  are decreasing in intensity relative to a line  $(L\gamma_1)$  which has an  $L_{II}$  initial state.

### VIII. CONCLUSIONS

The widths of lines arising from  $L_{\rm I}$  transitions increase strikingly with atomic number in the range  $73 \leq Z \leq 81$ . At the same time the intensities of these lines relative to lines arising from  $L_{II}$ and  $L_{III}$  transitions show corresponding decreases. These observations are accounted for qualitatively by the role of the Auger effect. Only two

Auger transitions are considered in the present discussion. Although beyond doubt these two are the principle ones involved in the topics of the present paper, there are certainly many more Auger transitions some of them with similar dramatic changes in probability from element to element. It is highly probable that many of the anomalies in x-ray line widths and relative intensities in other atomic number ranges will soon be shown to be due to Auger transitions.

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# Radiations from Radioactive Gold, Tungsten, and Dysprosium\*

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Level schemes are deduced for gold and tungsten from the measurement of beta-gamma and gamma-gamma coincidences, together with a knowledge of the energies of the various radiations emitted. The beta-ray spectrum of Au<sup>198</sup> is simple with an end-point of 0.78 Mev. The residual nucleus emits two gamma-rays in cascade. W187 has a complex beta-ray spectrum. The high energy group has a maximum energy of 1.4 Mev while the one of lower energy has an end-point of 0.5 Mev and is followed by a 0.9 Mev gamma-ray and some other gamma-rays which give gamma-gamma coincidences. The dysprosium beta-ray end-point is 1.2 Mev and the maximum energy gamma-ray is 1.1 Mev.

 $\mathbf{I}^{\mathrm{N}}_{\mathrm{radioactive nuclei, it is necessary to know}}$ the energies and intensities of all gamma-rays emitted during the process, the energy distribution of all beta-rays emitted, and also any correlations between beta-rays emitted by the parent nucleus and the gamma-rays produced by the product. Up to the present our knowledge of all these factors, for any radioactive species, has been quite limited. In general one has available, or can determine with ease, data on the maximum energy of the electrons emitted during the process. In some cases the shape of the betaray spectrum is also known and information is

available as to whether the spectrum is simple or complex. In those cases in which the spectrum is complex the value of the inner end point has not been determined with any great accuracy. Frequently gamma-rays have been found and the energies and intensities of these have been determined in a few instances.

Norling,<sup>1</sup> Mitchell, Langer and McDaniel,<sup>2</sup> and Dunworth<sup>3</sup> have applied coincidence counting methods to the latter problem and have been able to derive reasonable energy level schemes for several radioactive transformations. In particular, level schemes for the disintegration of Mn<sup>56</sup> (2.5 hours), In<sup>116</sup> (54 min.), Sb<sup>124</sup> (60 days), and others have been determined. It is the pur-

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