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The Differences Between the M_5 and M_4 Absorption Edges of Gold in the Pure Metal and in a Gold-Copper Alloy

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The suggestion that the discrepancies between observed and computed energies of the M_4 and M_5 absorption edges arise because transitions occur from the M_4 and M_5 levels to lattice levels, the transitions being governed by some selection rule, was investigated. The M_4 and M_5 edges of gold in the pure state and in a 50 atomic percent copper-gold alloy were photographed in a vacuum spectrometer. The change of lattice constant from 4.06A for gold to 3.86A for the alloy, which caused the energies of the lattice levels to change, produced shifts toward higher energies in Au M_5 of 2.5 electron volts and in Au M_4 of 6.8 electron volts. This shows that the final levels of these edge transitions are lattice levels. The results give credence to the assumption that, upon assigning to the low lattice levels the values of l which they have in free atoms, there are preferred atomic to low lattice level transitions which are often given by $\Delta l = \pm 1$. Data of other investigators are cited to support the view that the final levels in absorption are lattice levels and that the l selection rule is in evidence.

INTRODUCTION

MANY of the five M x-ray absorption edges of 20 of the heavier elements have been measured.[1–11] Their energy values may be calculated[12] if one has at hand measured values of the L edges and certain emission lines. However, the observed values of the M edges are not, in most cases, those calculated. In the M_4 and M_5 edges discrepancies occur in 14 of the 18 elements studied. Six of the $8M_1$ edges observed probably show small discrepancies while the M_2 and M_3 edges yield discrepancies only occasionally. The observed energy value of the edge is usually greater than the calculated value.[13]

No general statement relating the magnitudes of the M discrepancies to physical quantities such as atomic number, for example, can now be made. Chemical binding[8, 14] does not appear to affect the discrepancies.

Siegbahn[15] and Phelps[9] have proposed that the M discrepancies occur because atomic electrons may not all go to the same final level in an absorption edge transition. Phelps further suggested that the final levels are lattice levels.

In view of the above it seemed important to obtain more direct evidence, than had previously been given, showing whether or not the M edge transitions occur between atomic and lattice levels.

The energies of the lattice levels should change if the atomic spacing is changed. If the proposal of Phelps were correct, the edges of a metal in its pure state and in an alloy of different lattice constant should occur at different frequencies; the appearance of such a shift would be an indication that the M electrons show preferences as to their final levels in edge-giving transitions.[16]

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[1] H. W. B. Skinner and J. E. Johnston, Proc. Roy. Soc. **161**, 420 (1937).

[2] V. H. Sanner, Zeits. f. Physik **94**, 523 (1935).

[3] J. A. Prins and A. J. Takens, Zeits. f. Physik **84**, 65 (1933).

[4] C. A. Whitmer, Phys. Rev. **38**, 1164 (1931).

[5] R. V. Zumstein, Phys. Rev. **25**, 747 (1925).

[6] E. Lindberg, Zeits. f. Physik **54**, 632 (1929); **56**, 402 (1929); **57**, 797 (1929).

[7] R. A. Rogers, Phys. Rev. **30**, 747 (1927).

[8] A. J. M. Johnson, Phys. Rev. **34**, 1106 (1929).

[9] W. D. Phelps, Phys. Rev. **46**, 357 (1934).

[10] D. Coster, Phys. Rev. **19**, 20 (1922).

[11] W. Stenström, Dissert. Lund. (1919).

[12] Calculated values for the edges (i.e., for atomic energy levels) are given by M. Siegbahn, *Spektroskopie der Röntgenstrahlen*, pp. 336–337 and 346–351. No uniqueness is attached to the use of L edge energies for computation. The final levels for the L edge transitions are simply reference levels and the discrepancies noted are the energy differences between these levels and the final levels for the M edge transitions.

[13] For discrepancy magnitudes the following is a brief statement: M_1, the six discrepancies are between 9 and 25 electron volts; M_2 and M_3, only four discrepancies are fairly large and they lie between 13 and 20 ev; M_4 and M_5, 24 of the discrepancies are between 20 and 50 ev, the others being smaller.

[14] E. Lindberg, Zeits. f. Physik **50**, 91 (1928).

[15] M. Siegbahn, Zeits. f. Physik **67**, 567 (1931).

[16] If it were true that an M electron was jumping to one of the very lowest levels in a lattice energy zone, and if this zone had below it an absolutely forbidden energy region wider than the observed shift, then this shift would not indicate an electron preference. However, this situa-

The photographing of the M_4 and M_5 edges of a metal were undertaken to test this point.

The M edges were chosen for investigation because: (a) Shifts of a few volts could be more easily detected for edges at longer wave-lengths,[17] (b) It had been well established from fine structure data that the final levels in K and L edge transitions were lattice levels. This had not been shown for M edges.

The Method

The alloy to be investigated was chosen after a consideration of several factors: (1) At least one of the component metals should be of high atomic number so that the M edge wave-lengths would be short enough to be easily photographed. (2) The alloy must be a homogeneous solid solution. (3) The lattice of the alloy must be of a type as near that of the pure metal, whose edges were desired, as possible. (4) The lattice constant of the alloy must be significantly different from that of the pure metal. (5) To facilitate making the metal absorbing screens the vapor pressures of the two metals should be of the same order of magnitude and the two metals should be able to diffuse into each other readily.

The alloy chosen was a 50 atomic percent each copper-gold alloy. This percentage was chosen as being a probable optimum for the combination of a relatively large expected shift and ease of photography.

A 50 atomic percent copper-gold alloy has a face-centered lattice of atomic spacing 3.86A[18] as compared to that of 4.06A[19] for pure gold. The ordering temperature of this alloy is about 430°C and the ordered structure is face-centered tetragonal with the longer side 3.95A and the two shorter sides 3.66A.

The shifts expected on the basis of Coster and Veldkamp's[20] work with the gold L_3 edge in a 50–50 gold-copper alloy and on the Kronig theory, to be discussed later, were about 4 volts. It was, therefore, decided to study the M_5 and M_4 edges as they could be expected to give significant results with the apparatus used.

There is an optimum thickness for an absorber to give edge contrast. It can be shown that, if μ_1 and μ_2 are the linear absorption coefficients on the short and long wave-length sides, respectively, the maximum value of the ratio of the change in intensity at the edge to the incident intensity is given when the thickness, t, is:

$$t=\frac{\log_e(\mu_1/\mu_2)}{\mu_1-\mu_2}.$$

A similar formula may be derived for the edge of a metal in an alloy:

$$t=\frac{\log_e[(\mu_1'+k)/(\mu_2'+k)]}{\mu_1'-\mu_2'},$$

wherein μ_1' and μ_2' are the absorption coefficients of the edge producing alloyed metal on its short and long wave-length sides, respectively, while k is the absorption coefficient of the other metal at that wave-length.

The values of the absorption coefficients were obtained from Jönsson's[21] formula and tables of Jönsson's values[22] of the so-called electron absorption coefficient. The computed optimum thicknesses were: for the gold absorber; M_5, 4.1×10^{-5} cm, M_4, 2.6×10^{-5} cm; for the alloyed gold; M_5, 4.8×10^{-5} cm, M_5, 3.6×10^{-5} cm.[23] The screens used were from 2.5 to 4.5×10^{-5} cm thick.

The metal absorbing films were made by evaporation in a vacuum. The metals were evaporated from a trough-shaped molybdenum filament. The bases for the films were made of blown soft glass which was about 10^{-4} to 10^{-3} cm thick.

The thickness of the evaporated films was governed in the following manner. After choosing

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tion is not often the case so that it seems probable that a shift does indicate the above mentioned preference.

[17] A shift of 0.01 cm on the photographic plate is about the smallest that one may consider significant on this apparatus. This corresponds to a shift in energy for the Au M_5 edge of 1.9 ev and for the Au L_3 edge of 69 ev. No shift, upon alloying, was reported (reference 20) for the Au L_3 edge.

[18] C. H. Johansson and J. O. Linde, Ann. d. Physik **25**, 1 (1936).

[19] Int. Crit. Tab., Vol. 1, p. 340.

[20] D. Coster and J. Veldkamp, Zeits. f. Physik **74**, 191 (1932).

[21] A. Jönsson, Dissertation, Upsala, 1928.

[22] Tables are given in M. Siegbahn, *Spektroskopie der Röntgenstrahlen*, p. 470.

[23] C. L. Andrews, Phys. Rev. **54**, 994 (1938) has recently measured absorption coefficients of gold. From values taken from a graph given by him calculated optimum thicknesses came out about 30 percent greater. The experience of the author in this research indicates Andrew's values give a calculated thickness nearer the actual optimum.

a permanent filament to base distance, known amounts of gold were put in the filament and evaporated. Upon measuring the weight and area of the deposit the thickness was computed. The amount of gold to be put in the filament for various film thicknesses was then known. This was repeated for the gold-copper films.

To insure that the alloy films were 50 atomic percent of both gold and copper a charge of gold and copper in equal atomic amounts and of total weight to give the required thickness was put in the filament each time. All the metal was evaporated.

TABLE I. *Pure and alloyed gold M_5 and M_4 edges. The alloyed gold discrepancies are also calculated on the basis of the pure gold L edge energies. Presumably, the L edges of gold in a gold-copper alloy would also be shifted, although this has not been reported,*[17, 20], *but in view of the relative magnitudes of these M discrepancies and the probable smallness of the L edge shifts any such L shift could not greatly affect these M discrepancies.*

EDGE	PLATE	λ *A*	V‡ EV	ΔM EV	SHIFT IN ALLOY *A*	SHIFT IN ALLOY EV
Au M_5	31	5.524	2233.2	37.5		
	51	5.525	2232.8	37.1		
	67	5.520	2234.8	39.1		
	68	5.518	2235.6	39.9		
	69	5.521	2234.4	38.7		
	70	5.523	2233.6	37.9		
	Average	5.522	2234.1	38.4		
	Lindberg*	5.529	2231.2	35.5		
	Johnson†	5.506	2240.9	45.2		
Alloyed Au M_5	41	5.513	2237.5	41.8	−0.009	3.4
	42	5.516	2236.4	40.7	− .006	2.3
	46	5.518	2235.6	39.9	− .004	1.5
	48	5.514	2237.2	41.5	− .008	3.1
	49	5.509	2239.3	43.6	− .013	5.2
	50	5.518	2234.6	39.9	− .004	1.5
	63	5.519	2235.2	39.5	− .003	1.1
	64	5.520	2234.8	39.1	− .002	0.7
	65	5.514	2237.2	41.5	− .008	3.1
	66	5.513	2237.5	41.8	− .009	3.4
	Average	5.515	2236.6	40.9	− .007	2.5
Au M_4	55	5.320	2318.8	36.5		
	75	5.315	2321.0	38.7		
	76	5.319	2319.3	37.0		
	88	5.321	2318.4	36.1		
	90	5.316	2320.6	38.3		
	Average	5.318	2319.6	37.3		
	Lindberg*	5.330	2314.5	32.2		
	Johnson†	5.315	2321.0	38.7		
Alloyed Au M_4	77	5.302	2326.7	44.4	− .016	7.1
	78	5.305	2325.4	43.1	− .013	5.8
	80	5.301	2327.1	44.8	− .017	7.5
	Average	5.303	2326.4	44.1	− .015	6.8

* Lindberg's measurement. See reference 6.

† Johnson's values. See reference 8. Measurements of Johnson's photographs by this author give a value for M_5 of 5.519A and for M_4 5.323A. The presence of $W(N_4 \rightarrow M_2)$ on his plates made the location of M_4 difficult.

‡ Calculated values: M_5, 2195.7 volts; M_4, 2282.3 volts.

In order that the evaporated films might approximate homogeneity the filament temperature was held somewhere near 1450°C where the vapor pressures of gold and copper are about equal. The molybdenum content of the films was negligible because at that temperature the vapor pressure of molybdenum is about 10^{-6} that of gold or copper.

The alloy absorption screens were then raised to a temperature of about 450°C, in a hydrogen atmosphere, and held there for periods from one to six hours. They were oven-cooled at the rate of about 2.5°C per minute. Such a treatment would certainly make the alloys homogeneous and probably order them.[18, 24–26]

The self-rectifying tube and vacuum spectrometer used have been described by Zumstein[5] and by Rogers.[7] Only slight modifications were made. Voltages on the tube were about 4100 volts, currents of 100 to 200 milliamperes were used, and exposure times were from 5 to 50 hours, depending on whether the photograph was one of emission, pure gold absorption, or alloy gold absorption.

The wave-lengths of the edges were determined by measuring visually[27] with a traveling "one-power microscope" the distances between known emission lines and the edges on the photographs, and then interpolating with a formula given by Whitmer.[4] The reference lines were Au $M_{\beta 1}$(5.612A) and Mo $L_{\alpha 1}$(5.395A).[28]

RESULTS

In Table I are given the values of the edges in terms of wave-length and energy. The difference between the observed energy and calculated

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[24] G. D. Preston, J. Inst. of Metals **46**, 477 (1931).

[25] N. S. Kurnakow and N. W. Ageew, J. Inst. of Metals **46**, 481 (1931).

[26] O. Eisenhut and E. Kaupp, Zeits. f. Elektrochemie **37**, 466 (1931).

[27] This method of measuring the edge positions proved to be much superior to the use of the microphotometer which was probably due to the large photographic grain, relatively small edge contrast, and very small variations in edge straightness caused by random grain position and crystal face flaws. The microphotometer traces which were made were in good agreement with visual measurements. Another observer made measurements in good accord with those of the author.

[28] Reference 12, p. 477.

energy[12] is given as ΔM. The changes in the wave-lengths and energies of the gold M_5 and M_4 edges upon alloying the gold with the copper is given in the last two columns.

DISCUSSION

From the experience of others who have operated this spectrometer and a consideration of possible apparatus and measurement errors it was estimated that the measurements are accurate to within ± 2.2 volts (± 0.005A) for M_5, and ± 2.4 volts (± 0.006A) for M_4. Consideration also showed that the measurement of a shift in an edge would not be of less accuracy than the measurement of the edge alone for many experimental errors would tend to cancel out. It was judged that the shifts obtained—M_5, 2.5 volts; M_4, 6.8 volts—were statistically and physically significant.

A greater number of alloy gold M_4 edge photographs was not obtained due to the inordinate amount of time required to successfully photograph it. The M_4 shift is not regarded as being as definitely demonstrated as the M_5 shift but as corroborating the M_5 results.

Because of the nature of the effect of alloy composition on edge contrast and edge shift it appears that any inhomogeneity along the thickness dimension of the alloy absorber would result in an "average" edge giving a smaller shift than a homogeneous alloy. So that, for a perfectly homogeneous alloy, the M_5 shift would be somewhat higher than the 2.5-volt average. This is in good agreement with the relevant investigation of Coster and Veldkamp[20] and the shift to be expected if the final M_5 level is approximately "Kronig-like."[29]

Other investigators,[20, 30–33] working with K and L edges, have obtained evidence that, in the absorption process, atomic electrons go to lattice energy levels. No significant shift, of the nature already described, has been reported for these edges. However, there is observed an absorption fluctuating with wave-length on the short wave-length side of the edge. This absorption fine structure extends for several hundred volts from the edge. The fine structures of metals having the same lattice type have been shown to be similar while those from different lattice types are not similar. Metals in alloys were found[17, 27, 28] to have fine structures characteristic of the alloy lattice, and the component metals exhibited the same edge fine structure, thus showing that the fine structures are dependent on the lattice and not on the atoms themselves. This effect is observed[34] in the edges of elements in ionic crystals except that the fine structures of different anions must be compared and likewise those of the cations.[35]

All of the K and L edge data, so far obtained, show that for a given lattice type the voltage separations of the different maxima and minima of absorption from the main edge are inversely proportional to the square of the lattice constant.[36] The Kronig theory[37–39] has had much success in describing quantitatively the energy separations of these fine structures.[40] Kronig finds that allowed lattice electron energies are not restricted except for narrow forbidden zones whose energy values are inversely proportional to the square of the interplanar distance in the direction of electron motion.

In accord with these data, it has been found by Hanawalt[41] for Kr, Xe, Zn and Hg gases and vapors and by Coster and Van der Tuuk[42] for A

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[29] $\left[\frac{\text{lattice constant for Au}}{\text{lattice constant for alloy}}\right]^2 \Delta M_5(\text{Au}) = \Delta M_5\ (\text{alloy})$. $[4.06/3.86]^2\ 38 = 42$ volts. This indicates, roughly, a 4-volt shift in the M_5 edge.

[30] J. Veldkamp, Zeits. f. Physik **82**, 776 (1933).

[31] D. Coster and R. Smoluchowski, Physica **2**, 1 (1935).

[32] J. D. Hanawalt, Zeits. f. Physik **70**, 293 (1931).

[33] J. Veldkamp, Physica **2**, 25 (1935).

[34] G. A. Lindsay, Zeits. f. Physik **71**, 735 (1931).

[35] That an anion and cation in the same lattice show different fine structures is attributed to the fact that the two find themselves in different potential fields.

[36] Before this is evident, from the data, a small constant characteristic of the element and lattice must be added to the voltage separations of the maxima and minima from the main edge. This constant takes account of the fact that the initial electron level is characteristic of the atom and that the voltage separations from some reference level, not necessarily the final level for a transition giving the main edge, are the quantities which are inversely proportional to the square of the lattice constant.

[37] R. de L. Kronig, Zeits. f. Physik **70**, 317 (1931).

[38] R. de L. Kronig, Zeits. f. Physik **75**, 191 (1932).

[39] R. de L. Kronig, *Handbuch der Physik* **24½**, 290 (1933).

[40] Empirically, the fine structures within about 50 volts of the main edge are not quite so well described by his theory.

[41] J. D. Hanawalt, Phys. Rev. **37**, 723 (1931).

[42] D. Coster and J. H. Van der Tuuk, Zeits. f. Physik **37**, 367 (1926).

that their K and L edges show no fine structure more than the ionization potential of the atom beyond the main edge.

It appears beyond doubt that the K and L edge fine structures for solids are a result of electron transitions from atomic to lattice energy levels.

On the basis of the fine structure data it seems reasonable, then, to assume that the M electrons are also going to valence and lattice levels in edge transitions. Since the L edges themselves are caused by transitions to valence and very low lattice levels, and because of the magnitudes of the M discrepancies, it appears that usually the $M_{2,3}$ electrons[43] are jumping to valence and low lattice levels while the M_1, M_4, and M_5 electrons are going to higher lattice levels. Computations of the energies of the valence and very low lattice levels in a metal have been given.[44–46] In general, it may be said that these energy levels depend on the element, lattice type, and atomic spacing. Thus, the M edges, for which transitions occur to these low levels, should depend on these factors. Kronig's theory appears, empirically, to apply approximately to the energy region to which most M_4 and M_5 edge transitions take place; hence, it is useful as a guide.

The question of whether or not the ordinary x-ray selection rules ($\Delta l = \pm 1$, $\Delta j = \pm 1, 0$) are holding for atomic to low lattice level transitions is of importance. The description of the Kronig lattice levels depends in no way on a quantum number of the nature of l but while it does not appear directly in treating the valence and low lattice levels they certainly retain some of the free atom character which was described in part by the l quantum number.

Experimental evidence strongly indicates that the l selection rule does express the preferred atomic to valence and low lattice level transitions. There is little indication of a j selection rule. Sandström[47] found that the energy values of the L_2 and L_3 edges for Ta 73 to Au 79 are the same as those for the emission lines $O_4 \rightarrow L_2$ and $O_{4,5} \rightarrow L_3$ measured by Idei.[48] For these elements the $O_{4,5}$ levels are probably valence levels so that this is an indication that the l selection rule is giving the preferred transitions. Veldkamp[32] found that to match the fine structures of the L edges from the same metal for Ta, W, Pt, and Au the L_1 edge must be displaced to higher energies about 20 volts for Ta and W and about 15 volts for Pt and Au. This is definite evidence that the final level in the L_1 transition is above that for the $L_{2,3}$ transitions. From spectroscopic data and a knowledge of outer electron configurations in these elements Veldkamp concluded that the l selection rule was correctly indicating the transitions. The fact that the L_2 and L_3 edges of Ta and W show sharp line-like absorption he interprets as being due to a transition to their $5d$ shells.[49] The M_3 edges of Ta[4] and Os[7] have also been reported to be line-like. These data also indicate preferred transitions. Along this same line, the author has taken the difference between the M_1 and the (averaged) $M_{2,3}$ discrepancies for W, Os, Pt, and Au. He finds that they, too, are in close agreement with the spectroscopically determined $6p$ and $5d$ (or $6s$ for Pt and Au) energy differences.

There is correlation between the $M_{4,5}$ discrepancies and the first absorption minimum in the L_3 fine structure for the elements where comparison may be made. The comparisons indicate that the final levels to which the $M_{4,5}$ electrons go are not those which are preferred by the L_3 electrons.

All of these considerations indicate that, upon assigning to the lattice valence levels the values of l which they have in free atoms, there are preferred atomic to low lattice level transitions which are often given by a change in l of ±1. On this basis all edges involving the same l quantum number should show the same shift. It is not known that they do not; because of the small number of M_4 shifts measured the author is not certain that the difference in the observed M_4 and M_5 shifts is real.

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[43] This is apparent since the M discrepancies are all less than 50 volts and the main L edges themselves are caused by transitions to the valence and low lattice levels.

[44] E. Wigner and F. Seitz, Phys. Rev. **43**, 804 (1933).

[45] E. Wigner and F. Seitz, Phys. Rev. **46**, 509 (1934).

[46] J. C. Slater, Phys. Rev. **45**, 794 (1934).

[47] A. Sandström, Zeits. f. Physik **65**, 632 (1930).

[48] S. Idei, Science Reports, Tohoku Imp. Univ., Series I, **19**, No. 5.

[49] The $5d$ levels of these elements are rather sharp and have a high density of states.

Since it has been definitely shown in this and other investigations that the final levels in absorption edge transitions are lattice levels, and since the l selection rule is evidently giving the preferred transitions in many cases, it may be concluded that the M discrepancies arise because the M and L electrons may not all go the same level. One should expect, as has so far often been the case, M_1, M_4, and M_5 discrepancies but none for the M_2 and M_3 edges.[50]

For helpful assistance and counsel I wish to express my thanks to the staff of the Department of Physics of the State University of Iowa and especially to Professor G. W. Stewart who suggested the problem and directed the research.

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[50] If computations are made on the basis of the observed L_3 edge energy.

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New X-Ray Lines in the *L* Series Resulting from *K* Auger Transitions

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Three new x-ray lines in the L series of silver were found which have wave-lengths of 4.030A, 4.016A, and 3.805A. A fourth and much fainter line was found at 4.023A. These lines are due to processes in which an atom, initially ionized in the K shell, undergoes transitions of the type

$K\rightarrow LL+$expelled electron (Auger transition)
$LL\rightarrow LM+$quantum (radiative transition).

These lines differ from those previously observed from multiply ionized atoms in that they are more widely separated both from each other and from the diagram lines. An x-ray vacuum spectrograph capable of withstanding a potential of 100 kv across the x-ray tube was used in the observation of these lines. The anode of the x-ray tube was a thin foil of silver backed with aluminum. Special precautions were necessary to suppress the continuous background radiation in the region of the lines as the expected intensity of the lines was of the same order of magnitude as the intensity of the diagram line arising from the transition L_{III} to N_{I}.

INTRODUCTION

THIS paper presents the result of a search for some faint lines predicted in the L spectrum of silver.

A great deal of effort has been expended in the search for nondiagram lines in various atoms in an effort to learn more about atomic structure. Nondiagram lines were first observed by Siegbahn and Stenstrom[1] while they were studying the K series of the elements Na(11) to Zn(30), and have been reported by numerous others[2] a few of whom are listed in this paper.

Multiple ionization of the atom was assumed to explain the presence of these lines. Some of the early theories assumed that the atom was multiply ionized by a single electron impact while others assumed that the multiple ionization was due to successive ionizations by different impacting electrons. These theories were mainly at fault in that the predicted intensities were not in agreement with the experimentally observed intensities. Some of the theories attributed the lines to single transitions in multiply ionized atoms while others attributed them to double transitions in multiply ionized atoms.

Coster and Kronig[3] have improved the theory to account for the intensity of the L satellites by assuming an Auger[4] transition from L_{I} ionization to L_{III} with an accompanying M_{IV} or M_{V} ionization. This leaves the atom doubly ionized, which is the initial condition for the production of satellites. The number of atoms originally ionized in L_{I} is comparable to the number ionized in L_{III}

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[1] Siegbahn and Stenstrom, Physik. Zeits. **17**, 48 (1916); **17**, 318 (1916).

[2] G. Wentzel, Ann. d. Physik **66**, 437 (1921). D. Coster, Phil. Mag. **43**, 1070 (1922). A. Larsson and M. Siegbahn, Ark. f. Mat. Astron och Fysik, Stockholm **18**, 18 (1924). G. Wentzel, Zeits. f. Physik **31**, 445 (1925). L. A. Turner, Phys. Rev. **26**, 143 (1925). T. Wetterblad, Zeits. f. Physik **43**, 767 (1927). M. J. Druyvesteyn, Dissertation, Groningen (1928).

[3] D. Coster and R. de L. Kronig, Physica **2**, 13 (1935).

[4] P. Auger, J. de phys. et rad. **6**, 205 (1925).