EFFECT OF FIELD THEORETICAL CORRECTIONS ON X-RAY SPECTRA

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ABSTRACT

The wavelengths of K_{α_1}, K_{α_2}, K_{β_1}, K_{β_3}, K_{β_2}I and K_{β_2}II lines of heavy elements of atomic number 70–92 and the associated energy levels, K, L_{II}, L_{III}, M_{II}, M_{III}, N_{II} and N_{III} have been calculated using a modified form of the relativistic energy formula of Sommerfeld, incorporating in it both the total and internal screening constants in a closed form. All the field theoretical corrections in the second and the fourth order as well as the finite nuclear size correction have been included.

The relative intensities $K_{\alpha_2}/K_{\alpha_1}$, K_{β_3}/K_{β_1} and K_{β_2}II/K_{β_2}I have been calculated with relativistic wavefunctions. In addition to the retradation and screening, field theoretical corrections have also been used. A logical extension of the one electron-model with application of the field theoretical corrections improves the calculated results of not only the energy levels but also the relative intensities of X-ray lines.

INTRODUCTION

LAMB shift and other field theoretical corrections, so well known in the spectra of light elements,[1] have been made use of in the study of the energy levels and the relative intensities of X-rays lines. As the field theoretical corrections are inversely proportional to the cube of the principal quantum number of the level concerned and directly proportional to the fourth power of the atomic number, K-lines of the heavy elements of atomic number 70–92 have been chosen for this investigation. Part I gives the wavelengths of K_{α_1}, K_{α_2}, K_{β_1}, K_{β_3}, K_{β_2}I, K_{β_2}II lines and the associated energy levels and Part II describes the study of the relative intensities of these lines.

PART I

Energy Levels

In the one electron-model, X-ray energy levels are given by the Sommerfeld energy formula[2]

$$E(n, l, j) = Rhc \frac{M_Z}{M_Z + m}\left[\frac{(Z-\sigma_1)^2}{n^2} + \frac{\alpha^2(Z-\sigma_2)^4}{n^4}\left\{\frac{n}{j+\frac{1}{2}} - \frac{3}{4}\right\} + \frac{\alpha^4(Z-\sigma_2)^6}{n^6}\left\{\frac{1}{4}\left(\frac{n}{j+\frac{1}{2}}\right)^3 + \frac{3}{4}\left(\frac{n}{j+\frac{1}{2}}\right)^2 - \frac{3}{2}\left(\frac{n}{j+\frac{1}{2}}\right) + \frac{5}{8}\right\} + \ldots\right] \quad (1)$$

where σ_1 and σ_2 are the total and internal screening constants used by Sommerfeld.

In the calculations of L_I–L_{II} level separations, it was observed[3] that the agreement between the above formula and the experimental values is fortuitous as the higher terms are not negligible for heavy elements. It was also observed that the magnitude of Lamb shift and other field theoretical corrections is significantly large for heavy elements and it is essential to take them into consideration. When these corrections were incorporated in the relativistic formula with the necessary higher terms, agreement was once again obtained.

Modified Sommerfeld—Dirac Energy Formula

In the calculations of X-ray energy levels of heavy elements, since the higher terms in Sommerfeld's energy formula are not negligible, there is not very much justification for neglecting the higher terms. In fact, such a suggestion had come from Sommerfeld himself at a time when generally only the Z^2 term was taken.[4] But apart from the fact that an expansion in the form of a series to the required degree of accuracy will be tedious, truncation errors cannot be ruled out completely. Hence the usual Sommerfeld energy formual was modified by incorporating both the total screening constant, σ_1 as well as the internal screening constant, σ_2 in the closed formula as given by Dirac and Sommerfeld:[5]

$$E(n, l, j) = -\frac{1}{2}\frac{\alpha^2(Z-\sigma_1)^2}{n^2} + \frac{1}{2}\frac{\alpha^2(Z-\sigma_2)^2}{n^2} + \left\{1 + \frac{\alpha^2(Z-\sigma_2)^2}{[n' + \sqrt{k^2 - \alpha^2(Z-\sigma_2)^2}]^2}\right\}^{-\frac{1}{2}} \quad (2)$$

where $k = j + \frac{1}{2}$, and n' is given by $n = n' + k$.

FIELD THEORETICAL CORRECTIONS

The relativistic energy calculated by the above formula was corrected for Lamb shift or second order radiative correction, second order relativistic shift, second order magnetic moment correction, second order vacuum polarization correction, fourth order magnetic moment and vacuum polarization corrections, finite mass corrections and finite size corrections as given by Series.[6] The wavelengths of K_{α_1}, K_{α_2}, K_{β_2}, K_{β_1}, $K_{\beta_2 II}$ and $K_{\beta_2 I}$ X-ray lines and the associated energy levels, *viz.*, K, L_{II}, L_{III}, M_{II}, M_{III}, N_{II} and N_{III} levels were computed for elements of atomic number 70 to 92 as it was observed that the field theoretical corrections are large only for the heavy elements.

SCREENING CONSTANTS

Though originally Sommerfeld's screening constants had been derived from experimental data,[7] the screening constants themselves turned out to be correct, firstly because for elements of low atomic number, the higher terms in the Sommerfeld energy formula as well as the field theoretical corrections are negligible, and secondly, for heavier elements, the very omission of the higher terms inadvertently accounted for the above corrections.[3] Thus even though Lamb shift was unknown at that time, the screening constants stood the test of time. This was further checked by Pauling[5] whose values agree with the values of Sommerfeld in general.

Slater's screening constants,[8] which account for screening by inner electrons and derived independently of X-ray spectroscopic data, agree well with Sommerfeld's internal screening constants. In fact, though the Slater screening constants cannot replace the total screening constant σ_1, they are good substitutes for the internal screening constants, as shown by our calculations. This is another independent check for the essential correctness of Sommerfeld's internal screening constants.

Though the necessity for a complete evaluation of the correct screening constants cannot be ruled out, any attempt to recalculate the screening constants from experimental data will have to take Lamb shift and other field theoretical corrections into account.

Even among authors who have accepted Sommerfeld's internal and total screening constants, there seems to be a difference of opinion regarding the constants to be used for K level. Blokhin[2] takes 0·3 for σ_1 and zero for σ_2 for all elements. Sommerfeld[7] and Pauling[5] use a much higher value for

σ_1 of K level. This is also linearly dependent on Z. Pauling's calculated value for σ_2 is 0·169. In fact, as the X-ray spectra are due to one electron missing systems, a value of 0·3 is suggested for σ_2 of K level on the same analogy as Slater's screening.

RESULTS

Calculations have been made for K lines and associated energy levels with not only the modified Sommerfeld-Dirac energy formula with Lamb shift and other field theoretical corrections but also Dirac energy formula without any screening as well as with Slater screening. The different screening constants such as Sommerfeld's screening constants, Pauling's calculated values and Slater screening constants, in place of the internal screening constants as well as the different screening constants for the K level, were used. These results were compared with the experimental values given by Sandström[9] as well as Bearden[10] and Bearden and Burr.[11] Detailed calculations are given elsewhere.[12] The wavelengths of the K-lines of mercury (Hg 80) alone are given in Table I as an example. The calculated results given were obtained by the modified Sommerfeld-Dirac energy formula with Sommerfeld's screening constants with the field theoretical corrections; a value of 0·3 was used for σ_2 for the K-level. It is observed that the calculated values are nearer the experimental values of Sandström. As the differences between the experimental values themselves are large, the agreement obtained can be considered to be satisfactory. The assumption of the " Proton Halo " structure for the nucleus will increase the nuclear size corrections already calculated.[13, 14] However, Lamb shift and other corrections are considerably larger than finite size corrections. How exactly this type of structure will

TABLE I

Wavelengths of K-lines of mercury (Hg 80) in ν/R units

Values	K_{α_2}	K_{α_1}	K_{β_3}	K_{β_1}	K_{β_2}II	K_{β_2}i
Calculated ..	5076·343	5218·342	5889·034	5920·391	6082·765	6090·335
Observed (Sandström)	5075·000	5216·500	5882·200	5914·200	6075·100	6083·600
Observed .. (Bearden)	5063·771	5205·218	5866·992	5898·665	6058·957	6067·025

TABLE II

Calculated binding energies of K, L_{II}, L_{III}, M_{II}, M_{III}, N_{II} and N_{III} levels in ν/R units

Z	K	L_{II}	L_{III}	M_{II}	M_{III}	N_{II}	N_{III}
70	4524·379	744·597	668·080	164·305	148·051	30·718	27·089
71	4671·511	772·658	690·995	170·750	153·326	32·960	29·033
72	4821·587	801·409	714·320	177·367	158·707	33·449	29·207
73	4974·667	830·864	738·057	184·159	164·193	37·751	30·172
74	5130·788	861·038	762·208	191·131	169·786	36·361	31·426
75	5290·009	891·946	786·773	199·576	176·775	38·693	33·379
76	5452·369	923·605	811·756	208·255	183·918	41·110	35·395
77	5617·939	956·031	837·157	217·172	191·215	43·614	37·474
78	5786·764	989·241	862·979	226·331	198·667	46·208	39·617
79	5958·912	1023·254	889·222	235·738	206·275	48·891	41·825
80	6134·431	1058·089	915·890	245·397	214·041	51·667	44·097
81	6313·401	1093·764	942·983	255·315	221·965	54·536	46·434
82	6495·885	1130·302	970·504	265·496	260·047	57·500	48·837
83	6681·960	1167·724	998·454	275·946	238·291	60·562	51·306
84	6871·701	1206·051	1026·836	286·672	246·695	63·723	53·841
85	7065·183	1245·308	1055·652	297·679	255·262	66·985	56·444
86	7262·426	1285·520	1084·903	308·974	263·992	70·350	59·113
87	7463·637	1326·713	1114·592	320·563	272·888	73·820	61·851
88	7668·836	1368·913	1144·721	332·455	281·949	77·397	64·658
89	7878·143	1412·149	1175·291	344·655	291·176	81·084	67·533
90	8091·621	1456·451	1206·306	357·172	300·573	84·883	70·478
91	8309·446	1501·852	7237·767	370·014	310·138	88·796	73·493
92	8531·629	1548·383	1269·677	383·189	319·874	92·826	76·579

affect the radiative, vacuum polarization and other corrections is at present not known. But suffice to say that a large variation of the nuclear radius upto 10F as suggested by Barrett *et al.*[13] will even cause changes in the screening constants and thus affect the relativistic energy of the atoms. The above reasons or even other unknown effects which have not been taken into account might perhaps explain why the calculated values are only agreeing with Sandström's values and not Bearden's values though the latter values are more recent and accurate. The calculated values of binding energy for the K, L_{II}, L_{III}, M_{II}, M_{III}, N_{II} and N_{III} levels are given in Table II. The binding energy of K shell for mercury (Hg 80) calculated by different methods are given in Table III and compared with other available data.[15] This table shows that the modified Sommerfeld-Dirac energy formula with field theoretical corrections is able to give good values in comparison with other methods.

TABLE III

K ionization energy (in ν/R units) of mercury (Hg 80)

Experimental (Bearden's)	..	$6108{\cdot}06 \pm 0{\cdot}6$
Experimental (Sandström's)	..	$6121{\cdot}7$
Non-relativistic, Coulomb field ($Z = 80$)	..	6400
Relativistic, Coulomb field (Dirac)	..	7068
Relativistic with inner screening	..	7012
Relativistic Hartree-Fock (Coulthard)		6145 ± 8
Field theoretical and other corrections	..	57
With Pauling's screening constants in modified Sommerfeld—Dirac formula with field theoretical corrections	..	6139
With $0{\cdot}3$ for σ_2 for K-level and other calculations as before	..	6134

CONCLUSION

The general conclusions are given below:

(1) The values obtained from Dirac energy formula with and without Slater screening constants differ very much from the experimental values,

(2) Slater screening constants are, however, good substitutes for the internal screening constants.

(3) Values of 0·3 for the total screening constant and zero for the internal screening constants for the K-level as taken by Blokhin[2] do not give good results.

(4) Modified Sommerfeld-Dirac energy formula with field theoretical corrections gives results in good agreement with the experimental results.

PART II

RELATIVE INTENSITIES OF K-LINES

The problem of theoretically accounting for the relative intensities of the X-ray lines of heavy elements had been eluding solution for a long time. Recently, relativistic calculations of the intensities had been taken up by a number of authors with different degrees of refinements.[16–22] The oscillator strength and intensity ratios of the K-lines for elements of atomic number $Z = 70$ to 92 had been calculated by the authors earlier with not only retardation (as used by Payne and Levinger[19]) but also screening and field theoretical corrections.[23] These calculations showed that the use of relativistic energy with field theoretical and other corrections discussed in Part I improved the results when no screening was used. However, it was observed that when screening was used, application of the field theoretical corrections did not improve the results although as far as calculations of energy were concerned, such an application of the corrections to relativistic energy made agreement with experimental values much better.

It was shown by Scofield[22] that Babushkin's retardation formula was a better approximation than Payne's retardation formula, though it was expected that for heavier elements both the formulae might lead to the same results. Hence oscillator strengths and relative intensities were computed with Babushkin's retardation formula to study how far this would improve the results obtained from the one-electron model. Screening and field theoretical corrections were included as before.

The oscillator strengths for the transitions $1S \rightarrow nP_{1/2}$ and $1S \rightarrow nP_{3/2}$ according to Babushkin are given below:

$$f_{1S \rightarrow nP_{1/2}} = \frac{1}{2} \frac{mc^2}{\hbar\omega} | R_1 + R_2 - R_3 - 3R_4 |^2$$

$$f_{1S \rightarrow nP_{3/2}} = \frac{mc^2}{\hbar\omega} | R_1 + R_2 + 2R_3 |^2 \qquad (3)$$

where

$$R_1 = \int_0^\infty g_2 j_L(\omega r)\, g_1 r^2\, dr$$

$$R_2 = \int_0^\infty f_2 j_L(\omega r)\, f_1 r^2\, dr$$

$$R_3 = \int_0^\infty g_2 j_{L-1}(\omega r)\, f_1 r^2\, dr$$

$$R_4 = \int_0^\infty f_2 j_{L-1}(\omega r)\, g_1 r^2\, dr$$

where f_1, f_2, g_1, g_2 are the corresponding small and large components of Dirac wavefunctions and j_L and j_{L-1} are the spherical Bessel functions. L is the angular momentum of the photon given by

$$l + l' + L = 0 \pmod 2$$

i.e.,

sum $l + l' + L$ should be even.

The energy values obtained from different formulae such as Dirac's energy formula, with and without Slater screening constants, relativistic energy with field theoretical corrections as well as different screening constants in relativistic wavefunctions were used to study the effect of screening and energy on the relative intensities. Calculations showed that with Babushkin's retardation formula the variation in intensities was less when compared to Payne's formula. Furthermore, when field theoretical corrections were used in energy, significantly, the results improved with screening unlike the earlier results.

Comparison with Hartree-Fock Calculations

Scoffeld[22] had calculated the radiative decay rates for K and L levels with Babushkin's retardation and Hartree-Fock-Slater wavefunctions. A comparison of the results with the experimental values of Beckman[24] as well as Nelson and Saunders[25] showed that for K_{α_2} and K_{α_1} lines the intensity ratios calculated by Scoffeld agreed well with the values of Nelson and Saunders. These values were higher than the values of Beckman. However, for K_{β_2}/K_{β_1} intensity ratios, Scoffeld's values were decreasing with Z whereas the experimental values[26] were increasing with Z.

TABLE IV

Calculated and observed intensity ratios of $K\alpha_2/K\alpha_1$ lines

Z	1	2	3	4	5	6
70	0·555	0·542	0·535	0·502	..	0·572
71	0·557	0·544	0·536	0·503	..	0·574
72	0·559	0·546	0·538	0·504	..	0·576
73	0·562	0·548	0·540	0·504	0·537	0·578
74	0·564	0·550	0·542	0·505	0·539	0·579
75	0·567	0·552	0·544	0·505	0·541	0·581
76	0·570	0·555	0·546	0·506	0·543	0·583
77	0·573	0·557	0·548	0·507	0·546	0·584
78	0·576	0·560	0·550	0·508	0·548	0·586
79	0·579	0·562	0·553	0·508	0·550	0·588
80	0·582	0·565	0·555	0·509	0·553	0·589
81	0·585	0·568	0·558	0·510	0·555	0·591
82	0·589	0·571	0·560	0·511	0·557	0·592
83	0·593	0·574	0·563	0·512	0·560	0·594
84	0·596	0·578	0·566	0·513	0·562	0·595
85	0·601	0·581	0·569	0·515	0·564	0·596
86	0·605	0·585	0·573	0·516	0·567	0·598
87	0·609	0·588	0·576	0·517	0·569	0·599
88	0·614	0·592	0·579	0·519	0·572	0·600
89	0·618	0·596	0·583	0·520	0·574	0·601
90	0·623	0·600	0·587	0·522	0·577	0·602
91	0·628	0·605	0·591	0·523	0·579	0·603
92	0·634	0·609	0·595	0·525	0·582	0·604

Calculations are with modified Sommerfeld-Dirac relativistic energy with field theoretical corrections in frequencies of lines, and Dirac energy in wavefunctions, retardation formula of Babushkin and correction for ω, columns 1, 2, 3 and 4 are with zero,s Slater, Burns and Sommerfeld total screening constants. Columns 5 and 6 are least square fitted values of Beckman[24] and Nelson *et al*,[25]

Oscillator Strength and Energy Factor

The oscillator strength formula with retardation should correspond to the dipole approximation if multipole expansions are neglected. While comparing the oscillator strength formula of Payne with his revised one, Babushkin had shown in an earlier paper[21a] that

$$f_{nlj\to n'l'j'} = \frac{2m\omega}{3\hbar}\left[(2j'+1)(2l'+1)\,C^2(l'|l;\ 00)\right.$$
$$\left.\times W^2(l'j';\ \tfrac{1}{2}\ 1)\ |R_1+R_2|^2\right] \qquad (4)$$

for the dipole transitions.

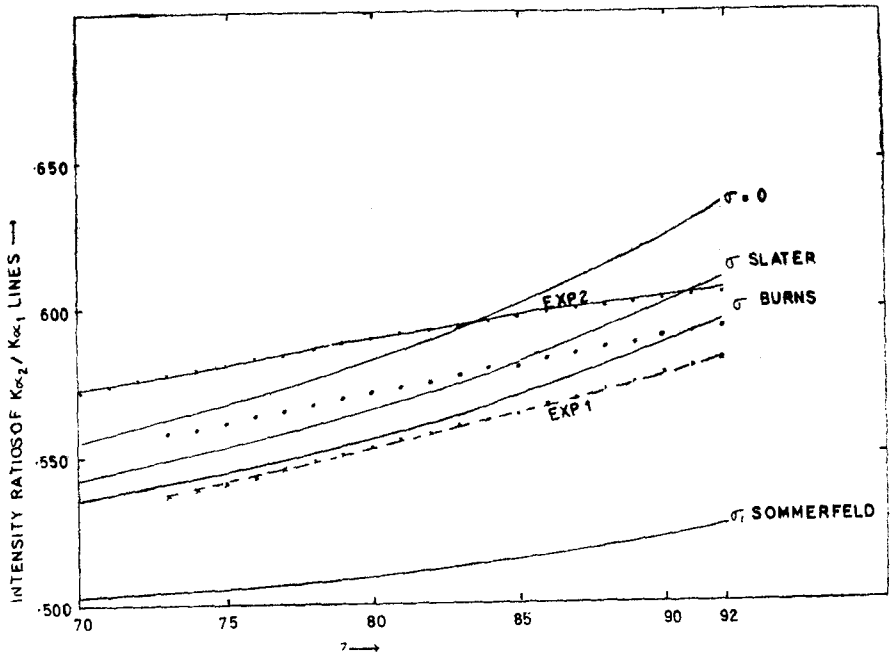

FIG. 1. Calculated intensity ratios of $K_{\alpha_2}/K_{\alpha_1}$ lines with field theoretical corrections, Babushkin's retardation formula and diffeeent screening constants in wave-functions and their comparison with experimental values. Exp. 1, curve fitted from Beckman's values and Exp. 2, from those of Nelson *et al.*, circles denote mean of Exp. 1 and Exp. 2.

Here the correction had not only been for radial matrix elements but also for the energy factor outside the square brackets, with ω in the numerator. In fact, the appearance of ω in the denominator of the oscillator strength formula will make intensity and transition probability formulae miss a factor of ω^2. Hence calculations were made with this correction factor

TABLE V

Calculated and observed intensity ratios of $K_{\beta 3}/K_{\beta 1}$ lines

Z	1	2	3	4	5	6
70	0·525	0·505	0·499	0·476	0·512	..
71	0·526	0·505	0·500	0·475	0·512	..
72	0·527	0·506	0·501	0·475	0·513	..
73	0·529	0·507	0·501	0·475	0·513	0·527
74	0·530	0·508	0·502	0·475	0·513	..
75	0·532	0·509	0·503	0·475	0·514	0·529
76	0·534	0·510	0·504	0·475	0·514	..
77	0·536	0·511	0·505	0·475	0·515	..
78	0·538	0·513	0·506	0·475	0·515	..
79	0·540	0·514	0·507	0·475	0·516	0·531
80	0·542	0·515	0·508	0·475	0·516	..
81	0·544	0·517	0·510	0·476	0·517	..
82	0·547	0·518	0·511	0·476	0·518	0·534
83	0·549	0·520	0·513	0·476	0·519	..
84	0·552	0·522	0·514	0·477	0·520	..
85	0·555	0·524	0·516	0·477	0·521	..
86	0·558	0·526	0·518	0·478	0·522	..
87	0·561	0·528	0·519	0·478	0·523	..
88	0·564	0·530	0·521	0·479	0·524	..
89	0·568	0·532	0·523	0·480	0·525	..
90	0·571	0·535	0·526	0·481	0·527	0·540
91	0·575	0·537	0·528	0·482	0·528	..
92	0·579	0·540	0·530	0·483	0·529	0·543

Calculations are with modified Sommerfeld-Dirac relativistic energy with field theoretical corrections in frequencies of lines, and Dirac energy in wavefunctions, retardation formula of Babushkin and correction for ω, Columns 1, 2, 3 and 4 are with zero, Slater, Burns and Sommerfeld total screeing constants. Columns 5 and 6 are least square fitted values of Beckman[24] and Salem, Seunders and Nelson.[26]

TABLE VI

Calculated intensity ratios of $K_{\beta 2}{}^{\prime\prime I}/K_{\beta 2}{}^{I}$ *lines*

Z	1	2	3	4
70	0·510	0·482	0·473	0·459
71	0·510	0·482	0·473	0·458
72	0·511	0·482	0·473	0·457
73	0·512	0·482	0·473	0·456
74	0·513	0·483	0·473	0·456
75	0·514	0·483	0·473	0·456
76	0·516	0·483	0·473	0·455
77	0·517	0·484	0·473	0·455
78	0·518	0·484	0·474	0·455
79	0·520	0·485	0·474	0·455
80	0·522	0·485	0·474	0·455
81	0·523	0·486	0·475	0·455
82	0·525	0·487	0·475	0·455
83	0·527	0·487	0·476	0·455
84	0·529	0·488	0·476	0·455
85	0·531	0·489	0·477	0·455
86	0·533	0·490	0·478	0·455
87	0·536	0·491	0·479	0·456
88	0·538	0·493	0·480	0·456
89	0·541	0·494	0·480	0·457
90	0·544	0·495	0·481	0·457
91	0·547	0·497	0·483	0·458
92	0·550	0·498	0·484	0·458

Calculations are with modified Sommerfeld-Dirac relativistic energy with field theoretical corrections in frequencies of lines, and Dirac energy in wavefunctions, retardation formula of Babushkin and correction for ω. Columns 1, 2, 3 and 4 are with zero, Slater, Burns and Sommerfeld total screening constants.

for ω^2 also. Screening, field theoretical corrections and retardation effects were taken into account as before. This correction factor had the effect of making the change of intensity ratios with Z less steep and the values were also less than those without this correction factor. A comparison of the different screening constants such as Slater's screening,[8] Sommerfeld's total screening constant[7] and Burns' screening[27] constant was made. Results are given in Tables IV, V and VI for the intensity ratios $K_{\alpha_2}/K_{\alpha_1}$, K_{β_3}/K_{β_1} and $K_{\beta_2}II/K_{\beta_3}I$ respectively. The values of $K_{\alpha_2}/K_{\alpha_1}$ and K_{β_3}/K_{β_1} are also shown in Figs. 1 and 2 for elements of atomic number 70 to 92. The results for $K_{\beta_2}II/K_{\beta_3}I$ are given in Fig. 3.

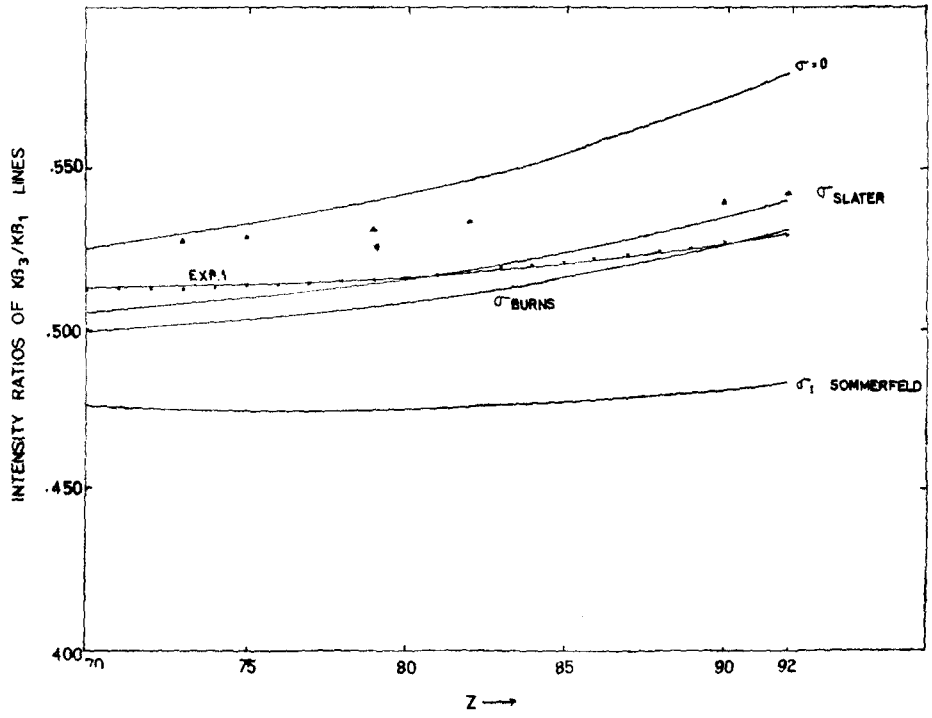

FIG. 2. Calculated intensity ratios of $K_{\beta 3}/K_{\beta 1}$ lines with field theoretical corrections, Babushkin's retardation formula and different screening constants in wavefunctions and their comparison with experimental values. Exp. 1, curve fitted from Beckman's values. Those obtained by Salem, Saunders and Nelson are shown as △.

When both $K_{\alpha_2}/K_{\alpha_1}$ as well as K_{β_3}/K_{β_1} intensity ratios are compared with the experimental results of Beckman as well as Nelson and Saunders, it is observed that with Slater screening constants, good agreement with experimental results is obtained.

The essential correctness of the calculated oscillator strength was further checked by a preliminary computation of the width of the lines as it is a more sensitive test for the oscillator strengths.

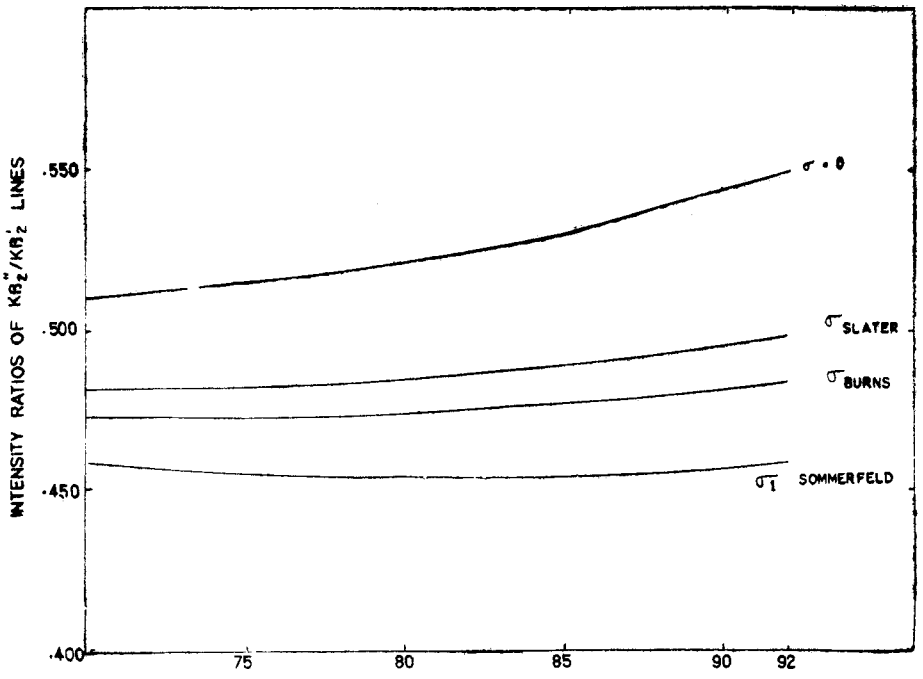

FIG. 3. Calculated intensity ratios $K_{\beta''_2}/K_{\beta'_1}$ lines with field theoretical corrections, Babushkin's retardation formula and different screening constants in wave functions.

These calculations have shown that a logical extension of the one-electron model by including field theoretical corrections such as Lamb shift gave not only the energies of K-lines in good agreement with experimental results but also the relative intensities of the lines of heavy elements.

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