MEASUREMENTS ON K X-RAY FLUORESCENCE YIELD RATIOS IN THE REGION OF $55 \leq Z \leq 82$

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ABSTRACT — Accurate measurements on the $K_{\alpha_2} / K_{\alpha_1}$, $K_{\beta'_1} / K_{\alpha_1}$, $K_{\beta'_2} / K_{\alpha_1}$, K_β / K_α and $K_{\beta'_2} / K_{\beta'_1}$, X-ray intensity ratios in elements for the region of $55 \leq Z \leq 82$ have been made using natural targets with ^{241}Am as exciting source of radiation. The beta and electron capture decaying isotopes were studied to measure the K_β / K_α ratios in some elements. High resolution Si (Li) and HpGe systems were used for the detection of X-rays. The experimental values of K_β / K_α are consistent with theoretical predictions due to Scofield, which include exchange corrections, except for $K_{\beta'_2} / K_{\beta'_1}$. A second degree polynomial fit to the experimental values of intensity ratios is found to be most suitable for the computation of K_β / K_α ratios.

1 – INTRODUCTION

In recent years, accurate measurements on K-X-ray intensities have assumed great importance in view of the availability of high resolution solid state detectors and fast computers. Simultaneously, refined theories have been proposed which permit a comparison of the experimental values with theoretical predictions. Several authors [1-6, 12] have measured the K X-ray transition rates in the region $55 \leq Z \leq 82$, employing crystal spectrometers as well as solid state detectors. They used different modes of excitation, namely photons, electrons and heavy ions as primary sources of radiation. Alternatively, the K X-ray intensities are measured via electron capture decay or internal conversion in radioactive isotopes. The K X-ray transition rates and K_β / K_α ratios measured using heavy ions are reported [7, 8] to be consistently higher than theoretical predictions due to Scofield [9]. It is also observed that this discrepancy between theory and experiment is found to be more pronounced as the charge of the bombarding ion increases. This is ascribed to the multiple ionisation of the atom in the case of heavy ions. Several authors have pointed out [1-6] that the K_β / K_α ratios determined via photoionisation caused by monochromatic X-rays or gamma rays, bremsstrahlung radiation and electron bombardment, are slightly higher than the Scofield theoretical values [9]. The original Scofield theory [9] assumes that the atomic electrons are in single particle states under the influence of a central potential given by the relativistic Hartree-Slater potential. Scofield modified his theory including exchange corrections [10] in the calculation in addition to the relativistic Hartree-Slater potential. The theoretical predictions of the modified Scofield theory [10] are seen to be in good agreement with the experimental K_β / K_α ratios. It may be noted that none of the earlier studies [1-6] on K_β / K_α ratios covers all the elements of the region considered here. Also, the available experimental results are due to different experimental techniques. A comparision of the predictions due to the modified Scofield theory [10] is made using these experimental values. This has prompted us to undertake a systematic experimental study of as many elements as possible in the region $55 \leqslant Z \leqslant 82$ employing high resolution solid state detectors with a computer facility, under identical conditions of experimentation, using ^{241}Am as the exciting source. In the case of some electron capture and beta decaying isotopes, a Hyper pure Germanium (HpGe) detector was used. The present work also covers the intensity ratios of several elements for which there were no previous measurements. The concerned elements are those with $Z = 55$, 57, 60, 65, 71 and 75. The values of the intensity ratios of K_β and K_α components are reported in this paper, compared to the theoretical predictions and discussed.

2 – EXPERIMENTAL DETAILS

The experiments were carried out in two phases. Under Phase I, a ^{241}Am source was used to create primary vacancies in the K-shell of an atom. Under Phase II, the intensity ratios were measured directly using radioisotopes. These isotopes are ^{137}Cs (beta), ^{133}Ba (EC), ^{152}Eu (EC, beta), ^{153}Gd (EC), ^{170}Tm (EC, beta), ^{185}W(beta), ^{203}Hg (beta), ^{204}Tl (EC, beta) and ^{207}Bi (EC). A high resolution fluorescence spectrometer was used for the detection and measurement of characteristic K X-rays. A block diagram of the present system including the source-target-detector geometry is shown in Fig. 1. The system includes a Si (Li) detector operated at liquid nitrogen temperature and coupled to a cooled FET preamplifier. It is also provided with a computer based 4K multichannel analyser and software programmes to process X-ray spectra and determine the final characteristic X-ray inten-

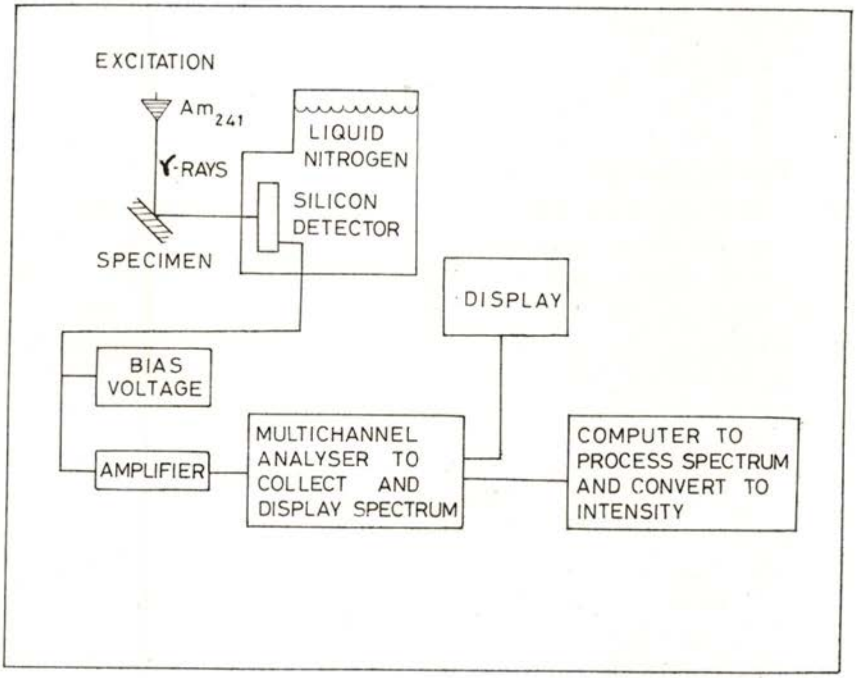

Fig. 1 — Block diagram of the experimental set-up

sities. The exciting source of radiation is a sealed 30 mCi ^{241}Am radioisotope. The resolution of the detector was measured to be 160 eV at 5.9 keV line of ^{55}Mn. Some of the radioisotopes were studied using a HpGe spectrometer facility of Bhabha Atomic Research Centre (BARC), Bombay, India. The energy resolution of the HpGe spectrometer was measured to be 180 eV at 5.9 keV. The resolution of the present systems is, thus, seen to be high enough to split close lying X-ray lines. The manipulation of the X-ray spectrum was done by a built-in computer of the type RT-11 with 32 K memory, using suitable software programmes.

3 – EFFICIENCY CALIBRATION

To determine X-ray intensities with the present detectors, a careful efficiency calibration is important. The low energy lines of well calibrated ^{57}Co, ^{133}Ba and ^{241}Am sources were used for the Si (Li) and HpGe spectrometers. The calibration was checked for the K/L ratios of X-rays following internal conversion in ^{137}Cs and ^{203}Hg.

4 – SOURCE-TARGET-PREPARATION

The natural targets like La, Ce, Pr, Nd, Tb, Dy, Ho and Lu were taken in their oxide form. They were pressed uniformly between two mylar films of thickness 0.00024 inches. The thickness of these samples ranges from 150 to 200 μg/cm^2. The radioactive sources ^{137}Cs, ^{133}Ba, ^{152} Eu, etc. of the present study were prepared by allowing a drop of the corresponding isotope on a mylar film to evaporate to dryness. A drop of insulin was also added to each source, so as to ensure an uniform spreading of the source material.

5 – DATA COLLECTION

The natural targets of different elements were excited by the 59.5 keV gamma rays from the 30 mCi ^{241}Am source. The characteristic X-ray spectra from different elements were recorded over long time intervals so as to ensure good statistics ($<$ .1 %). One typical characteristic X-ray spectrum from 'Ho' is shown in Fig. 2.

The figure shows that all the K X-ray components are well resolved. Fig. 3 shows the X-ray spectrum following electron capture decay of ^{207}Bi, as recorded by a HpGe system of BARC, Bombay, India. The intensities of X-ray lines were calculated using computer fits as well as by hand, after defining the

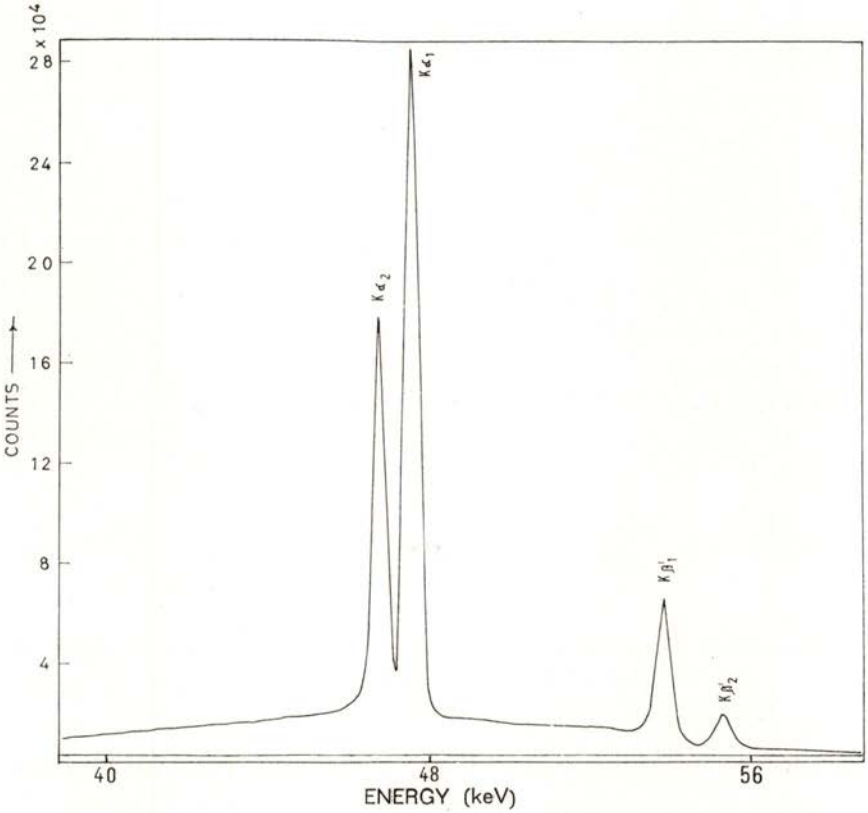

Fig. 2 — X-ray fluorescence spectrum from Ho recorded with a Si (Li) detector

background level. The two methods gave consistent values within 1 %. The areas thus obtained were corrected for self absorption and absorption in the air path between target and detector. Finally, they were corrected for the photopeak efficiency of the system used, in each case. The errors in the experimental values were estimated taking into consideration: 1) counting statistics,

2) instrument instability, 3) efficiency correction and 4) background correction. The experimental values of different ratios of K-components are summerized in Tables 1 through 5. The same tables

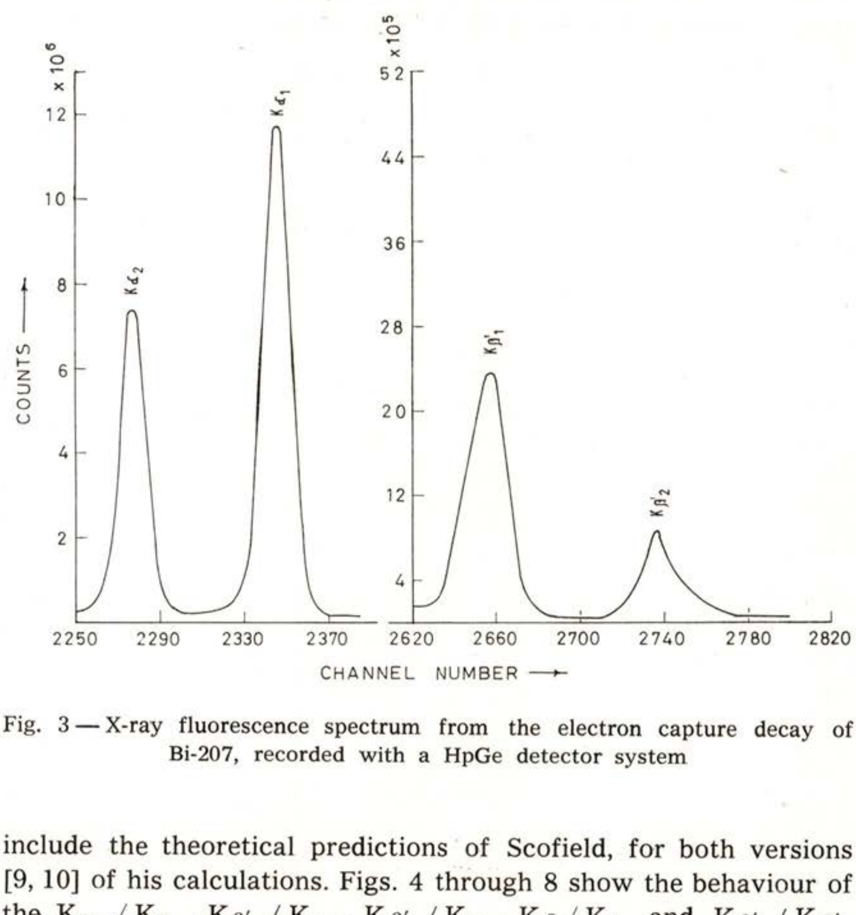

Fig. 3 — X-ray fluorescence spectrum from the electron capture decay of Bi-207, recorded with a HpGe detector system

include the theoretical predictions of Scofield, for both versions [9, 10] of his calculations. Figs. 4 through 8 show the behaviour of the $K_{\alpha_2} / K_{\alpha_1}$, $K_{\beta'_1} / K_{\alpha_1}$, $K_{\beta'_2} / K_{\alpha_1}$, K_{β} / K_{α} and $K_{\beta'_2} / K_{\beta'_1}$ ratios as functions of Z. The predictions due to Scofield [9, 10] are also shown in the same figures.

6 — RESULTS AND DISCUSSION

A polynomial fit was made to the experimental values of $K_{\alpha_2} / K_{\alpha_1}$, $K_{\beta'_1} / K_{\alpha_1}$, $K_{\beta'_2} / K_{\alpha_1}$, K_{β} / K_{α} and $K_{\beta'_2} / K_{\beta'_1}$ as a function of Z, for several degrees of freedom. The computations were

TABLE 1 — The experimental and theoretical values of the X-ray intensity ratios, $K_{\alpha_2} / K_{\alpha_1}$

Element	Z	Experimental values	Scofield theoretical values	
			Old, Ref. [9]	New, Ref. [10]
Cs	55	0.538 ± 0.011	0.542	0.542
Ba	56	0.540 ± 0.011	0.543	0.543
La	57	0.542 ± 0.011	0.545	0.544
Ce	58	0.544 ± 0.011	0.546	0.546
Pr	59	0.544 ± 0.011	0.548	0.548
Nd	60	0.545 ± 0.011	0.549	0.549
Sm	62	0.553 ± 0.012	0.553	0.553
Eu	63	0.551 ± 0.011	0.554	0.554
Gd	64	0.562 ± 0.011	0.556	0.556
Tb	65	0.559 ± 0.011	0.558	0.558
Dy	66	0.563 ± 0.011	0.560	0.560
Ho	67	0.559 ± 0.011	0.562	0.562
Er	68	0.560 ± 0.012	0.564	0.564
Yb	70	0.570 ± 0.011	0.567	0.567
Lu	71	0.569 ± 0.011	0.570	0.570
Re	75	0.576 ± 0.011	0.578	0.578
Hg	80	0.591 ± 0.011	0.590	0.590
Tl	81	0.594 ± 0.011	0.593	0.593
Pb	82	0.596 ± 0.011	0.595	0.595

TABLE 2 — The experimental and theoretical values of the X-ray intensity ratios, $K_{\beta'_1} / K_{\alpha_1}$

Element	Z	Experimental values	Scofield theoretical values	
			Old, Ref. [9]	New, Ref. [10]
Cs	55	0.294 ± 0.009	0.281	0.297
Ba	56	0.296 ± 0.009	0.283	0.300
La	57	0.296 ± 0.009	0.286	0.302
Ce	58	0.294 ± 0.009	0.288	0.304
Pr	59	0.295 ± 0.009	0.291	0.306
Nd	60	0.295 ± 0.009	0.293	0.309
Sm	62	0.300 ± 0.010	0.298	0.313
Eu	63	0.302 ± 0.009	0.300	0.315
Gd	64	0.305 ± 0.009	0.302	0.317
Tb	65	0.309 ± 0.009	0.304	0.319
Dy	66	0.310 ± 0.009	0.306	0.320
Ho	67	0.313 ± 0.009	0.308	0.322
Er	68	0.319 ± 0.010	0.310	0.324
Yb	70	0.327 ± 0.009	0.314	0.327
Lu	71	0.328 ± 0.009	0.315	0.329
Re	75	0.342 ± 0.009	0.323	0.335
Hg	80	0.342 ± 0.009	0.331	0.343
Tl	81	0.342 ± 0.009	0.333	0.344
Pb	82	0.342 ± 0.009	0.334	0.346

TABLE 3 — The experimental and theoretical values of the X-ray intensity ratios, $K_{\beta'_2} / K_{\alpha_1}$

Element	Z	Experimental values	Scofield theoretical values	
			Old, Ref. [9]	New, Ref. [10]
Cs	55	0.069 ± 0.003	0.065	0.073
Ba	56	0.071 ± 0.003	0.068	0.076
La	57	0.073 ± 0.003	0.070	0.077
Ce	58	0.077 ± 0.003	0.070	0.077
Pr	59	0.080 ± 0.003	0.071	0.078
Nd	60	0.083 ± 0.003	0.072	0.079
Sm	62	0.088 ± 0.004	0.073	0.081
Eu	63	0.088 ± 0.003	0.074	0.081
Gd	64	0.090 ± 0.003	0.076	0.083
Tb	65	0.090 ± 0.003	0.076	0.083
Dy	66	0.090 ± 0.003	0.076	0.083
Ho	67	0.091 ± 0.003	0.077	0.084
Er	68	0.090 ± 0.004	0.077	0.084
Yb	70	0.090 ± 0.003	0.078	0.085
Lu	71	0.088 ± 0.003	0.080	0.087
Re	75	0.088 ± 0.003	0.085	0.093
Hg	80	0.097 ± 0.003	0.093	0.100
Tl	81	0.101 ± 0.003	0.094	0.102
Pb	82	0.105 ± 0.004	0.096	0.104

TABLE 4 — The experimental and theoretical values of the X-ray intensity ratios, K_β / K_α

Element	Z	Experimental values	Scofield theoretical values	
			Old, Ref. [9]	New, Ref. [10]
Cs	55	0.236 ± 0.006	0.224	0.240
Ba	56	0.238 ± 0.006	0.227	0.243
La	57	0.239 ± 0.007	0.230	0.245
Ce	58	0.240 ± 0.007	0.232	0.247
Pr	59	0.243 ± 0.007	0.234	0.249
Nd	60	0.245 ± 0.007	0.235	0.250
Sm	62	0.250 ± 0.007	0.239	0.253
Eu	63	0.251 ± 0.007	0.241	0.255
Gd	64	0.253 ± 0.007	0.243	0.257
Tb	65	0.256 ± 0.007	0.244	0.258
Dy	66	0.256 ± 0.007	0.245	0.259
Ho	67	0.259 ± 0.007	0.246	0.260
Er	68	0.260 ± 0.007	0.248	0.261
Yb	70	0.265 ± 0.007	0.250	0.263
Lu	71	0.266 ± 0.007	0.252	0,265
Re	75	0.273 ± 0.008	0.258	0.271
Hg	80	0.276 ± 0.008	0.266	0.279
Tl	81	0.278 ± 0.008	0.268	0.280
Pb	82	0.280 ± 0.008	0.270	0.282

TABLE 5 — The experimental and theoretical values of the X-ray intensity ratios, $K_{\beta'_2} / K_{\beta'_1}$

	Z	Experimental values	Scofield theoretical values	
			Old, Ref. [9]	New, Ref. [10]
Cs	55	0.235 ± 0.011	0.230	0.244
Ba	56	0.240 ± 0.011	0.238	0.252
La	57	0.247 ± 0.011	0.245	0.253
Ce	58	0.262 ± 0.012	0.243	0.254
Pr	59	0.271 ± 0.012	0.244	0.256
Nd	60	0.281 ± 0.012	0.245	0.257
Sm	62	0.293 ± 0.012	0.247	0.258
Eu	63	0.291 ± 0.012	0.248	0.258
Gd	64	0.295 ± 0.012	0.251	0.263
Tb	65	0.291 ± 0.012	0.248	0.259
Dy	66	0.290 ± 0.012	0.249	0.260
Ho	67	0.291 ± 0.012	0.249	0.260
Er	68	0.282 ± 0.012	0.249	0.260
Yb	70	0.275 ± 0.012	0.250	0.261
Lu	71	0.280 ± 0.012	0.253	0.264
Re	75	0.257 ± 0.011	0.264	0.277
Hg	80	0.284 ± 0.013	0.280	0.295
Tl	81	0.295 ± 0.013	0.283	0.297
Pb	82	0.307 ± 0.014	0.288	0.302

made using a programme 'polyfit'. It was found that a quadratic fit yields the minimum chi-squared value. The coefficients of the polynomial are given in Table 6; the corresponding curves are shown as solid lines in figures 4-8. These figures show that the theoretical fits are consistent with the experimental behaviour of the K X-ray intensity ratio as a function of Z. The theoretical values of the intensity ratios of K components for elements not covered in the present study are given in Table 7; this table includes also the experimental values. From Table 7, it can seen that agreement between theory and the experimental values is satisfactory.

The intensity ratio measurements in elements Nd, Tb, Lu and Re were made for the first time in the present investigation; the same applies also to the $K_{\beta'_2} / K_{\beta'_1}$ ratios. It may be seen from Fig. 4, that there is a good agreement between the experimental values of $K_{\alpha_2} / K_{\alpha_1}$ and both versions of the theory due to Scofield.

TABLE 6 — Coefficients of the second degree polynomial fit to experimental values of the K X-ray intensity ratios

Coefficients	$K_{\alpha_2} / K_{\alpha_1}$	$K_{\beta'_1} / K_{\alpha_1}$	$K_{\beta'_2} / K_{\alpha_1}$	K_β / K_α	$K_{\beta'_2} / K_{\beta'_1}$
A_0	0.471351×10^{0}	0.133785×10^{0}	-0.108496×10^{0}	0.244827×10^{-1}	-0.250858×10^{0}
A_1	0.624179×10^{-3}	0.318455×10^{-2}	0.486874×10^{-2}	0.525117×10^{-2}	0.145837×10^{-1}
A_2	0.110417×10^{-4}	-0.720098×10^{-5}	-0.284165×10^{-4}	-0.261357×10^{-4}	-0.977562×10^{-4}

TABLE 7 — Values of K_β / K_α corresponding to the polynomial fit and experimental values (when available) for some elements

Atomic Number	Interpolated Values	Experimental values			
		Ref. [3]	Ref. [11]	Ref. [2]	Ref. [1]
61	0.248	—	—	—	—
69	0.260	—	—	—	—
72	0.267	0.269 ± 0.008	—	—	—
73	0.269	—	—	0.269 ± 0.014	0.262 ± 0.005
74	0.270	0.271 ± 0.008	—	—	—
76	0.273	—	—	—	—
77	0.274	—	—	0.275 ± 0.014	—
78	0.275	—	—	—	0.271 ± 0.005
79	0.276	0.276 ± 0.008	0.276 ± 0.02	0.272 ± 0.014	—

From Figs. 5-7, it may be noted that the experimental values of $K_{\beta'_1} / K_{\alpha_1}$, $K_{\beta'_2} / K_{\alpha_1}$ and K_β / K_α agree better with the theoretical values of Scofield when exchange corrections are included. However, one might note that the deviations that show up in figures 5 and 6 combine to give the disagreement in the

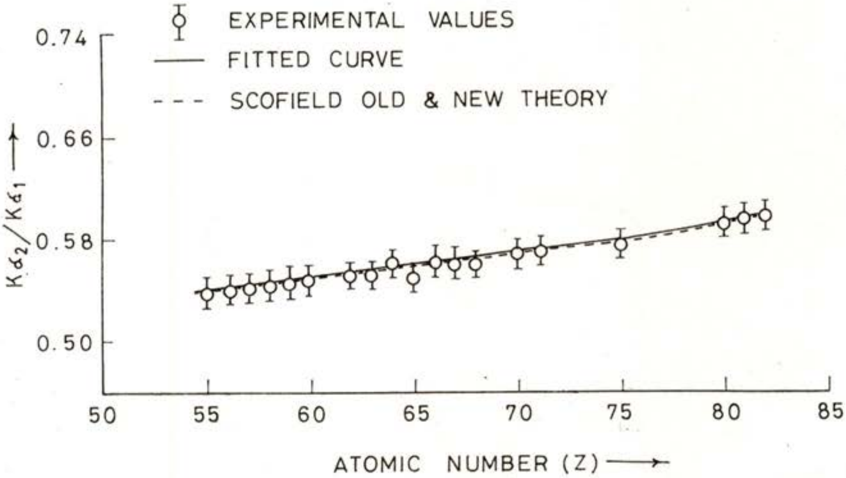

Fig. 4 — Experimental and theoretical curves showing $K_{\alpha_2} / K_{\alpha_1}$ versus atomic number Z

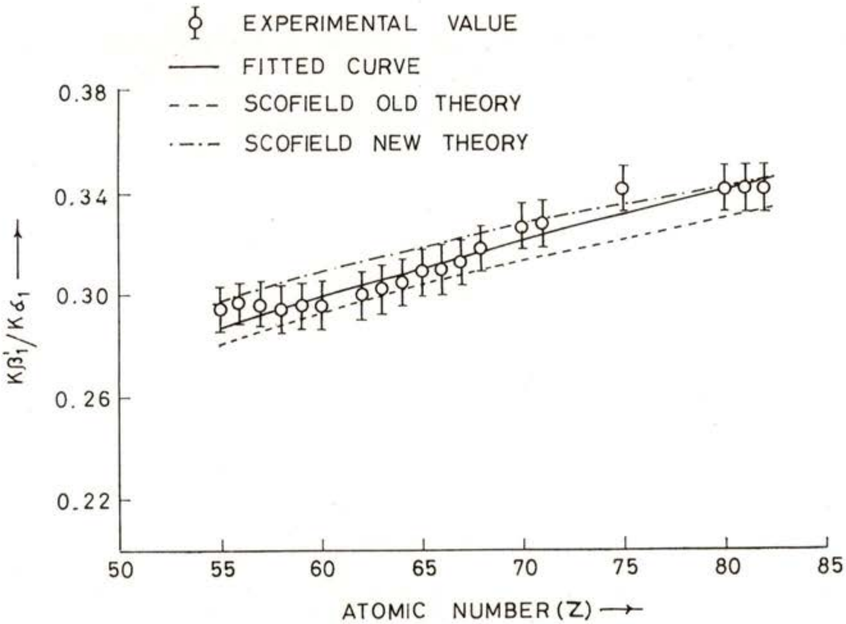

Fig. 5 — Experimental and theoretical curves showing $K_{\beta'_1} / K_{\alpha_1}$ versus atomic number Z

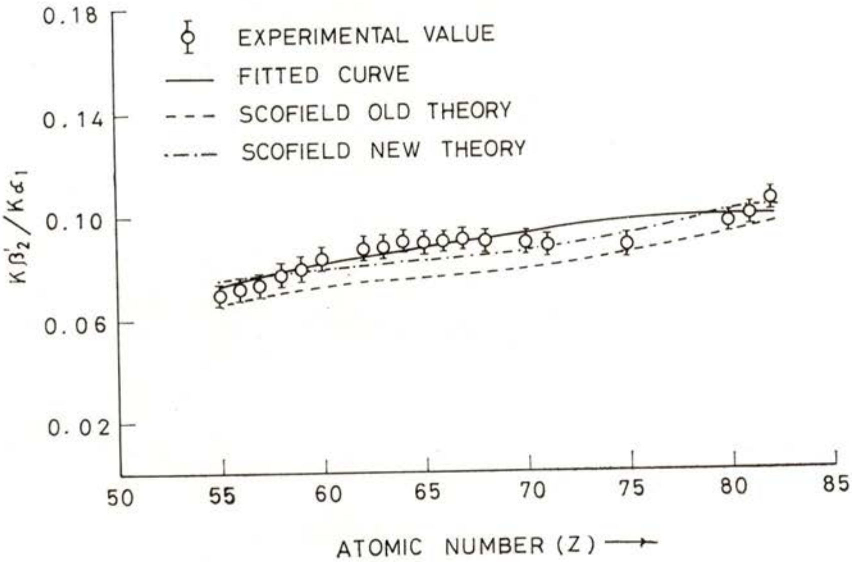

Fig. 6 — Experimental and theoretical curves showing $K_{\beta'_2} / K_{\alpha_1}$ versus atomic number Z

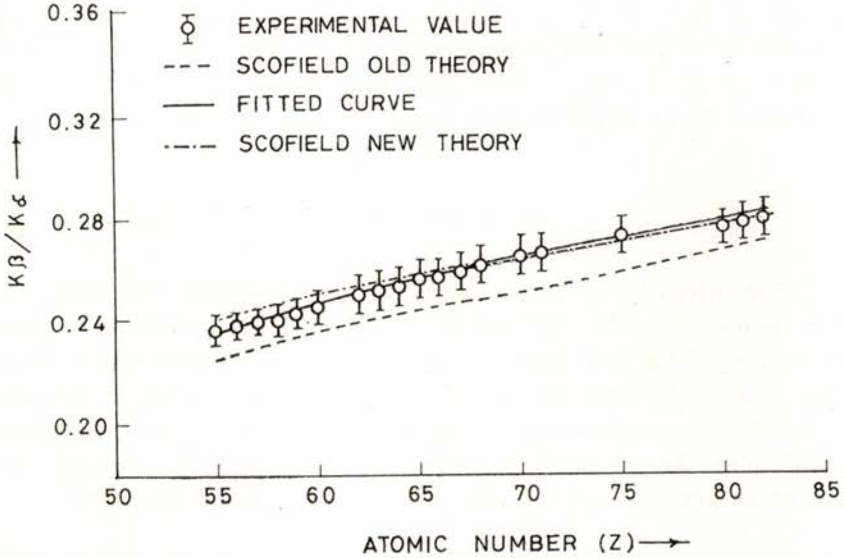

Fig. 7 — Experimental and theoretical curves showing K_{β} / K_{α} versus atomic number Z

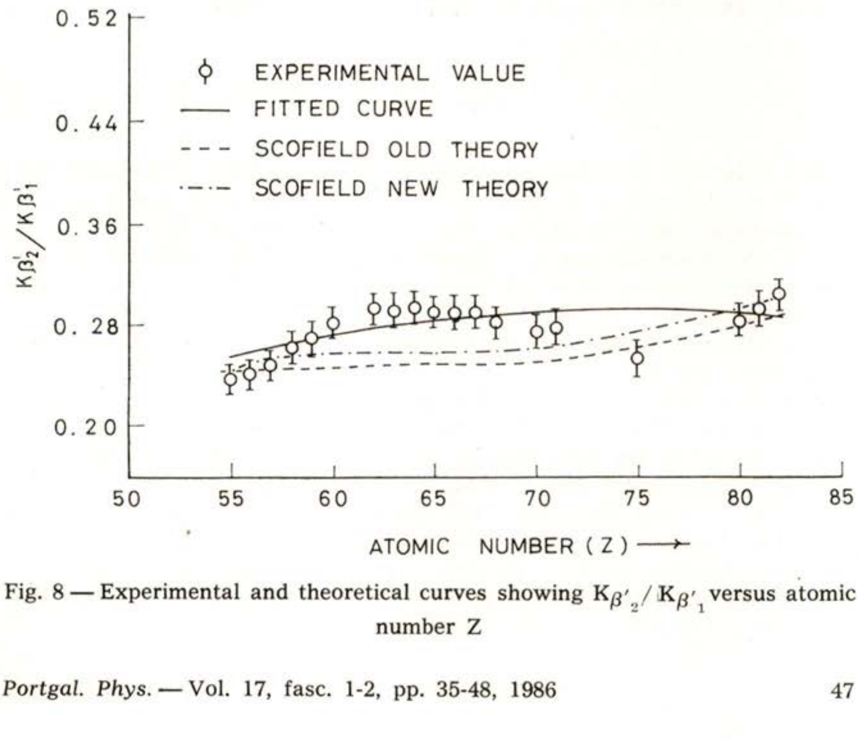

Fig. 8 — Experimental and theoretical curves showing $K_{\beta'_2} / K_{\beta'_1}$ versus atomic number Z

values of $K_{\beta'_2} / K_{\beta'_1}$ as evidenced in Fig. 8. Thus, it seems that the theory of Scofield is insufficient to explain the experimental values of $K_{\beta'_2} / K_{\beta'_1}$. One also sees from figure 8 that the quadratic fit to experimental points is not satisfactory.

7 — CONCLUSION

The intensity ratios of the K X-rays in different elements for the region $55 \leqslant Z \leqslant 82$, were measured using natural targets excited by 59.5 keV gamma rays of ^{241}Am. Radioisotopes were also studied to measure the K X-ray intensity ratios in certain elements. A comparison of experimental values and theoretical predictions due to Scofield shows that the exchange corrections are necessary to explain many of the observed K_β / K_α ratios.

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