Chapter 5

L-subshell ionization studies of some heavy atoms $73 \leq Z \leq 83$) with 59.54 keV photons

5.1 Introduction

Present chapter deals with the L-subshell ionization studies of some high Z elements ($73 \leq Z \leq 83$) by 59 54 keV photons The inner-shell vacancy is produced due to the interaction of the photon on the target electron Photon-atom interaction being a weak process, the production of multivacancy is less probable than in the case of ion-atom collision process It is appropriate to check the reliability of various atomic decay yield parameters available in the literature by photoionization studies

5.2 Relative L-Shell X-ray studies in Pt, Pb and Bi

5.2.1 Introduction

The photoionization phenomenon has been studied since very beginning of the century Studies of the L-shell X-ray fluorescence cross-sections of many elements and their intensity ratios have been reported in the past[1-7] Accurate determination of L-shell X-ray fluorescence cross-sections and their intensity ratios for different elements is important because of their wide use in the fields of atomic, molecular and radiation physics and in non-destructive elemental analysis of materials using either traditional photon sources or synchrotron radiation[8]

L-shell X-ray fluorescence cross-sections and their relative intensities can be calculated by using photoionization cross-sections, fluorescence yields, Coster-Kronig factors and partial radiative widths As the X-ray relative intensities can be determined with greater accuracy than the absolute cross-sections we have tried to measure the former and compared with the theoretical calculations The measurements were carried out in Pt, Pb and Bi using 59 54 keV γ-rays from an ^{241}Am point-source

5.2.2 Experimental Results and Discussion.

Our measured L_i X-ray intensity ratios along with the previously reported experimental results of Shatendra et al [2] and the theoretical results are presented in table-5 1 The theoretical results are calculated using the photoionization cross-sections of Scofield[9] given in table-5 2, and the decay yield data (see table-3 5) available in literature The partial radiative widths are taken from Campbell and Wang[10] A least-square fit to a typical experimental data is shown in fig 5 1 As is seen from table-5 1, our results for the L-shell X-ray intensity ratios of Pb and Bi are in good agreement with the previously published results of Shatendra et al [2] However, our present results are more accurate The L_i X-ray intensity ratios for Pt are reported for the first time and are found to lie between the two theoretical results obtained by using two sets of decay yield data The decay yield data of Werner and Jitschin[11] gives lower ratios for $\frac{I_\beta}{I_\alpha}$ and $\frac{I_\gamma}{I_\alpha}$ as compared to our experimental results. The decay yield data of Xu[12] and Chen et al [13] gives somewhat higher ratios for $\frac{I_\gamma}{I_\alpha}$, whereas, the ratios $\frac{I_l}{I_\alpha}$ and $\frac{I_\beta}{I_\alpha}$ are in good agreement.

Comparison of our results for Pb and Bi with theory suggests that the results obtained by using the decay yield data of Xu and Xu[14] are somewhat higher for the ratios $\frac{I_\beta}{I_\alpha}$ and $\frac{I_\gamma}{I_\alpha}$ In our earlier proton ionization studies[15] also the ratios obtained by using the decay yield data of Xu and Xu[145] were found to be higher than those obtained by using Krause's[16] decay yield data We therefore feel that the earlier conclusion of Xu and Xu[14] that their decay yield data are better than those of Krause[16] is not true

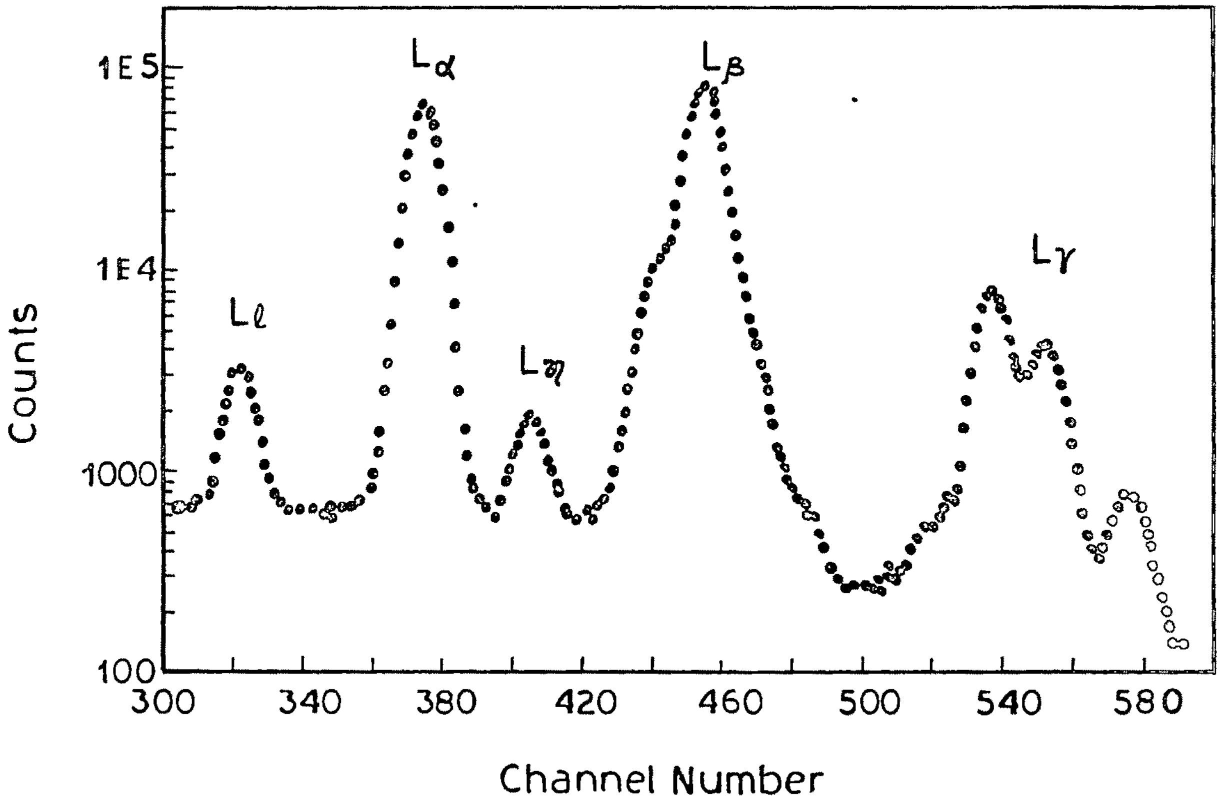

Fig 5 1 A typical L X-ray spectrum of Pb by 59 54 KeV photon impact

Table-5 1 Relative L-Shell X-ray Intensities with respect to L_α of Pt ,Pb and Bi following ionization by 59 54 keV photons

Target (Z)	Photon Energy(keV)	I_{L_l}/I_{L_α} Expt	I_{L_l}/I_{L_α} Theo	I_{L_β}/I_{L_α} Expt	I_{L_β}/I_{L_α} Theo	$I_{L_\gamma}/I_{L_\alpha}$ Expt	$I_{L_\gamma}/I_{L_\alpha}$ Theo
Pt^{78}	59 54	0 0491	0 0497[a]	1.121	1 183[a]	0 217	0 251[a]
		±0 0015		±0.038		±0 007	
			0 0496[b]		1 023[b]		0 231[b]
Pb^{82}	59 54	0.0517	0 0537[c]	1 073	1 032[c]	0 217	0 228[c]
		±0 0015		±0 025		±0 006	
		0 051[e]	0 0537[d]	1 203[e]	1 115[d]	0 235[e]	0 257[d]
		±0 004		±0 104		±0 022	
			0 0537[f]		1 211[f]		0 283[f]
Bi^{83}	59 54	0.0557	0 0536[c]	1 099	1 062[c]	0 234	0.230[c]
		±0 0015		±0 038		±0 007	
		0 062[e]	0 0536[d]	1 074[e]	1 127[d]	0 230[e]	0.252[d]
		±0.005		±0 098		±0 024	
			0 0536[f]		1 248[f]		0 286[f]

Table-5 2 Theoretical photoionization cross-sections[9] for K-shellof Cu and L-Subshells of Pt,Pb and Bi at 59 54 keV photon impact

Element (Z)	Photon Energy(keV)	σ_K	σ_{L_1} (in barns)	σ_{L_2}	σ_{L_3}
$_{29}Cu$	59 54	127 97			
$_{73}Ta$	59 54		344.03	170 01	197 2
Pt^{78}	59 54		420 25	247 95	274 87
$_{79}Au$	59 54		436.3	266 5	292 5
Pb^{82}	59 54		485 86	328 91	351 07
Bi^{83}	59 54		502 6	352 2	372.38

(a)Using the decay yield data of Xu[12] and Coster-Kronig data of Chen et al [13]

(b)Using the decay yield data of Werner & Jitschin [11]

(c)Using the decay yield data of Krause[14]

(d)Using the decay yield data of Xu and Xu[14] and Coster-Kronig factors of Werner and Jitschin[11]

(e)Experimental data of Shatendra et al [2]

(f)Using the decay yield data of Xu and Xu[14] and Coster-Kronig factors of Krause[16]

5.3 L-subshell ionization studies in Ta, Au and Pb:

5.3.1 Introduction

Measurement of L X-ray production cross-sections in heavy elements using a photon source are important from the point of view of their application in elemental analysis of materials The results of such studies can also be used for ascertaining the reliability of the available decay yield data sets For quantitative analytical applications it is necessary to know the relative intensities of different X-ray lines that contibute to the fluorescence In addition to their analytical applications, these measurements serve to provide a check on the theoretical calculations of some of the fundamental physical parameters, such as L-Subshell ionization cross-sections[9], fluorescence yields[8,11-14] Coster-Kronig factors[111-13] and radiative decay rates, the direct determination of which present many difficulties

Although X-ray tube sources have the advantage of large flux and offer monochromatic photons, the radio isotopes have the advantage of stable intensity and energy and of small size, which allows compact and efficient geometry.

Previous studies on some of the high Z elements($73 \leq Z \leq 92$) by Shatendra et al [2] using 59 54 KeV photons showed good agreement with the theoretical calculations but a recent study by Rao et al [7] using lower photon energies from secondary targets excited by K X-rays from a Tungsten X-ray tube- source showed consistently higher cross-sections than the theory The present study was undertaken for checking the theoretical predictions and adequacy of the presently available decay yield data sets Recently Xu and Xu[14] have made a detailed study on the decay yield parameters of Pb and Bi through a comparison of experimental L_i X-ray production cross-sections and their relative intensities induced by protons and α-particles with theoretical predictions As the heavy charged particle excitation data may be plagued with the multiple ionization processes it will be quite appropriate to use the photoionization data for such comparisons. The present data is hoped to serve that purpose

5.3.2 Experimental Results and discussion

The measured values of L_i X-ray production cross-sections of Ta, Au and Pb are listed in table-5 3 The X-ray production cross-sections are calculated from the theoretical L-subshell ionization cross-sections of Scofield[9] and different decay yield data sets(see table-3 5) The L-subshell ionization cross-sections are calculated from the measured X-ray production cross-sections with the help of an intricate iteration procedure which has been discussed earlier in the experimental chapter The fitting of L_γ line into four components (L_{γ_5}, L_{γ_1}, $L_{\gamma_{236}}$ and $L_{\gamma_{44'}}$) is shown in fig 5 2 and the residuals of the fitting is shown in fig 5 3

The errors quoted in table-5 3 are statistical only The errors coming from various corrections will be within 7% It is evident from table-5 3 that the experimental X-ray production cross-section values are, in general, higher than the theoretical predictions except for Au where the agreement seems to be better This may be because of availability of more accurate decay yield data for Au which had been measured by Jitschin et al [17] using synchrotron radiation For the other cases we find some random agreements for certain lines reflecting the inaccuracy of certain parameters in the decay yield data set The present data for Ta shows higher values for all the lines compared to the results of Shatendra et al [2,6] It is also to be noted that the theoretical estimations for Ta listed in table-1 of the paper of Shatendra et al [2] differ from our present results We feel that this discrepancy could be because of use of different radiative widths or may be because of some error in their numerical calculations Our present results of Ta for the individual and total cross-sections are about 25% higher than the theory This large discrepancy cannot be accounted as due to uncertainties in the corrections. It cannot be also due to improper normalization for the source intensity as we find reasonably good agreement for the Au data An accurate set of fluorescence yields and Coster-Kronig parameters will be useful for making a comment on the adequacy of the theoretical ionization cross-sections of Ta

The experimental X-ray production cross-sections of Au are in good agreement with the theoretical results obtained by using the decay yield data of Jitschin et al [17] The theoretical results corresponding to other decay yield data also provide reasonably good agreement with the data ,the Krause's data giving lower results for L_α and the theoretical decay yield data of Chen et al giving lower values for L_β

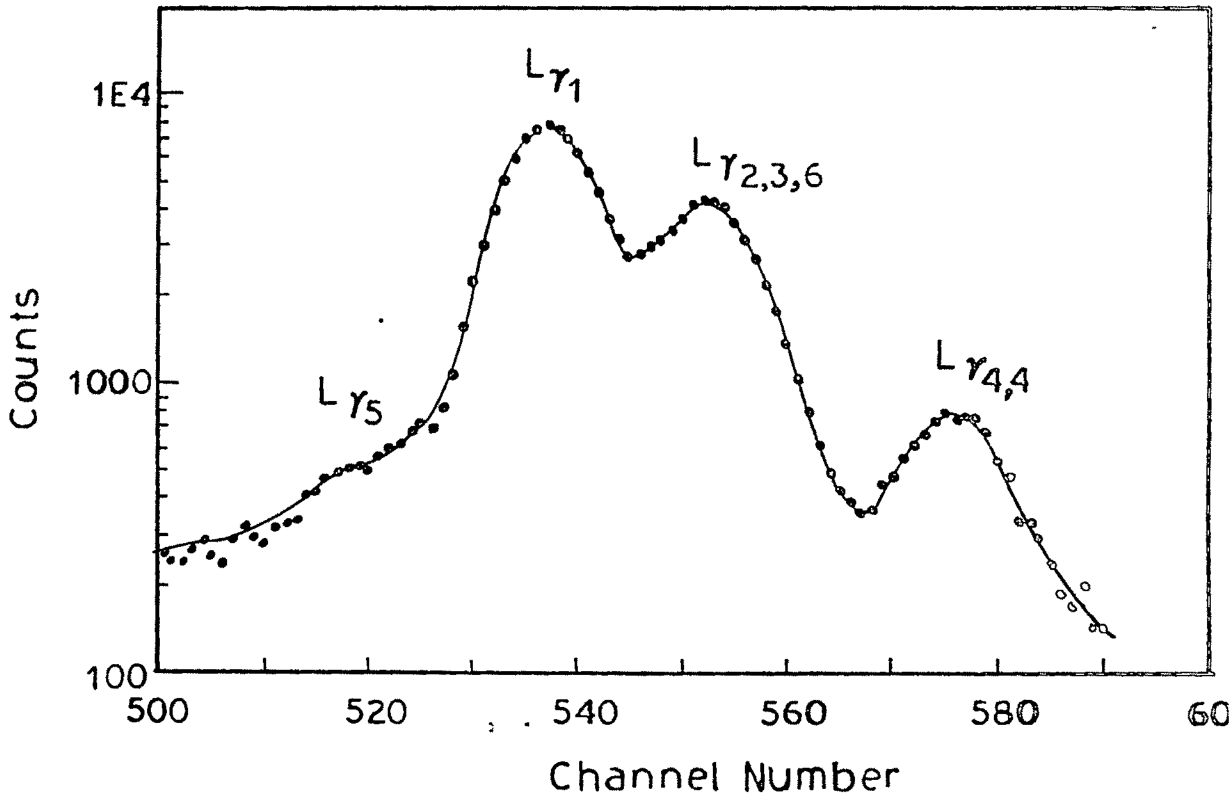

Fig 5 2 Measured(•) and fitted(—) L_γ spectrum of Pb

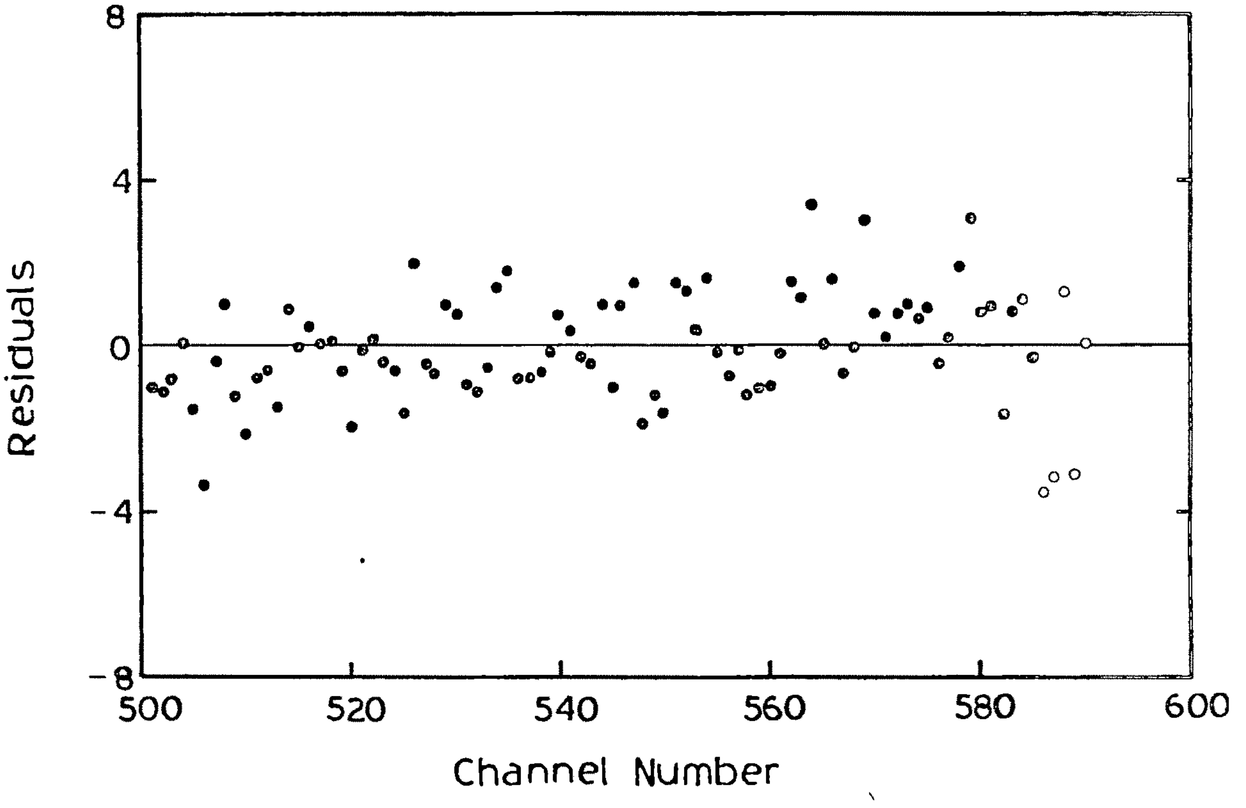

Fig 5 3· Residuals in units of one standard deviation of measured and fitted data of Pb L_γ X-ray peak and L_γ

In the case of Pb the experimental results for the X-ray production cross-sections are again higher(within 20%) than the theoretical results obtained from four different decay yield data sets The decay yield data of Xu and Xu[14] gives maximum discrepancy for L_α and L_γ The discrepancy in the total X-ray production cross-section is lowest for the decay yield data of Werner and Jitschin[6] The decay yield data of Xu and Xu[14] with the Coster-Kronig factors of Krause[16] gives similar agreement as that of Krause[16] The lower value of L_α cross-section corresponding to Xu and Xu's decay yield data[12] seems to be responsible for giving higher relative intensities with respect to L_α which is similar to the one reported by them for proton bombardment[12]

The measured relative intensities with respect to L_α are listed in table-5 4 The relative intensities for Ta and Pb are in better agreement with theory although the X-ray production cross-sections show large discrepancies This suggests that more accurate decay yield data are needed for both Ta and Pb for ascertaining the actual cause for the discrepancy in the X-ray production cross-sections As is seen from table-5 4 the relative intensities for Ta by Shatendra et al [2] are much higher than present results whereas their results for Pb are in good agreement with ours The present results for the relative intensities of Pb are also in good agreement with our earlier results[18] presented in the previous section The relative intensities for Au agree reasonably well with the theoretical predictions as well as with the experimental results of Shatendra et al[2]

The measured ionization cross-sections are compared with the theoretical results of Scofield[9] in table-5 5 Large discrepancies are found in the total ionization cross-sections of all the samples The agreement in the individual ionization cross-sections fluctuates depending upon the decay yield data set σ_{L_2} and σ_{L_3} deviate more for Ta whereas for Au the decay yield data of Jitschin et al [17], provides somewhat better agreement For Pb, the decay yield data of Krause[16] gives higher discrepancy for σ_{L_1} whereas the other decay yield data[11,12] give good agreement For σ_{L_2} the decay yield data of Xu and Xu[14] gives better agreement For σ_{L_3} the decay yield data of Xu and Xu[14] gives the maximum deviation of 45%, whereas the decay yield data of Kiause[16] gives better agreement

From the measured X-ray production cross-sections we have estimated the fluorescence yield parameteis ω_1 and ω_2 using the equations (3 8) and (3 11) and Coster-Kronig factor f_{12} of Krause[16] for Ta and Pb and of Jitschin et al.[17] for Au In this estimation we have subtracted the contribution of L_{γ_6} intensity from $L_{\gamma_{236}}$ by taking $\frac{\tau_{\gamma_6}}{\tau_{\gamma_1}}$ ratio from Campbell and Wang[10] and the measured intensity $I_{L_{\gamma_1}}$ We have also estimated the average L-fluorescence yields $\bar{\omega}_L$ using the relation

$$\sigma^x_{L_T} = \bar{\omega}_L \sigma^I_{L_T} \tag{5 1}$$

where $\sigma^x_{L_T}$ is the sum of the measured x-ray production cross-sections $\sigma^x_{L_l}$, $\sigma^x_{L_\alpha}$, $\sigma^x_{L_\beta}$, and $\sigma^x_{L_\gamma}$ and $\sigma^I_{L_T}$ is the total theoretical L-shell ionization cross-section which has been taken from Scofield[9] These are presented in table-5 6 The measured ω_1 and ω_2 parameters of Ta are found to be higher than the reference data of Krause[16] Our ω_1 and ω_2 results of Au are in good agreement with Jitschin et al [17] Our value of ω_1 for Pb, is found to be in good agreement with the data of Krause[16] but lower than that of Xu and Xu[14] whereas, ω_2 is higher than the value given by Krause[16] but is in close agreement with that of Xu and Xu The average L-fluorescence yield parameter $\bar{\omega}_L$ is found to be consistently higher($\sim$15%) than the recently reported data of Hubbell et al.[19]

We have also estimated the value of ω_3 in all the cases by taking the Coster-Kronig factors of Krause[16] for Ta and Pb and of Jitschin et al [17] for Au using the experimental ω_1 and ω_2 values obtained as mentioned above and the ionization cross-sections of Scofield[9] in the expression for L_α X-ray production cross-section and equating it with the measured cross-section(eqn 3 6) The ω_3 values so obtained are also given in table 5 6. Our ω_3 results are found to be somewhat higher than the presently available results.[14,16,17]

Table-5 3 Comparison of experimental X-ray production cross-sections (in barns) with the theoretical estimations obtained by using Scofield's ionization cross-sections and different decay yield data sets The errors quoted are statistical only The error due to sample self absorption, detector efficiency and air absorption will be within 7%

Target	$\sigma^X_{L_l}$		$\sigma^X_{L_\alpha}$		$\sigma^X_{L_\beta}$	
Atom(Z)	Theory	Expt	Theory	Expt	Theory	Expt
$_{73}Ta$	3.13[a]	4 3±0 2	67 5[a]	87 5±0 9	95 6[a]	120 1±1 2
		2 7±0 2[g]		58±4[g]		115±6[g]
$_{79}$Au	7 36[a]	8.7±0 4	145 8[a]	174 5±1 6	153 5[a]	179.7±1 7
	7 50[b]	7.7±0 6[g]	148 6[b]	146±9[g]	167 8[b]	154±7[g]
	8 05[c]		159 6[c]		144 0[c]	
$_{82}$Pb	10 36[a]	11.5±0 60	193 1[a]	226 1±2 2	199 2[a]	237 9±2 4
	9 38[d]	9 6±0 5[g]	174 8[d]	187±12[g]	211 8[d]	225±13[g]
	9 68[e]		180 4[e]		201 1[e]	
	10 27[f]		191 4[f]		208 1[f]	

Table 5 3 continued

Target	$\sigma^X_{L_{\gamma_{15}}}$		$\sigma^X_{L_\gamma}$		$\sigma^X_{L_T}$	
Atom(Z)	Theory	Expt	Theory	Expt	Theory	Expt
$_{73}$Ta	9 05[a]	13 36 ±0 26	20 6[a]	27 1 ±0 6	186 6[a]	239 ±2 4
				23±2[g]		198 7±7 5[g]
$_{79}$Au	17.82[a]	20 06 ±0 37	30 0[a]	34 1 ±0 6	336 7[a]	397 1 ±3 7
	18 74[b]		33 4[b]	34[g] ±2	357 5[b]	341 7[g] ±11 6
	17 27[c]		26 7[c]		338.4[c]	
$_{82}$Pb	25 1[a]	29 57 ±0 60	43 91[a]	52.0 ±1 1	446 6[a]	527 5 ±5 3
	27 2[d]		49 6[d]	44[g] ±3	445 6[d]	465 6[g] ±18
	24 5[e]		46 5[e]		437 6[e]	
	24 6[f]		48 0[f]		457 8[f]	

(a) Krause[16] (b) Jitschin et al [17] (c) Chen et al [13] (d) Xu and Xu[14]+Krause[16] (e) Xu and Xu[14]+Werner and Jitschin[11] (f) Werner and Jitschin[11] (g) Shatendra et al [2]

Table-5 4 Comparison of experimental L Shell x-ray production cross-section ratios with respect to L_α with the theoretical estimations obtained by using Scofield's ionization cross-sections[9] and different decay yield data sets

Target	$\frac{\sigma^X_{L_l}}{\sigma^X_{L_\alpha}}$		$\frac{\sigma^X_{L_\beta}}{\sigma^X_{L_\alpha}}$		$\frac{\sigma^X_{L_\gamma}}{\sigma^X_{L_\alpha}}$		$\frac{\sigma^X_{L_T}}{\sigma^X_{L_\alpha}}$	
Atom(Z)	Theory	Expt	Theory	Expt	Theory	Expt	Theory	Expt
$_{73}Ta$	0 0464[a]	0 0488 ±0 0015	1 416[a]	1 372 ±0 019	0 305[a]	0 310 ±0 009	2 76[a]	2 73 ±0 04
		0 0465[g] ±0 0047		1 983[g] ±0.171		0 397[g] ±0 044		
$_{79}$Au	0 0505[a]	0 0502 ±0 0015	1 053[a]	1 030 ±0 015	0 206[a]	0 195 ±0 006	2 31[a]	2 28 ±0 04
	0 0505[b]		1 029[b]		0 225[b]		2 41[b]	
	0 0505[c]	0 0527[g] ±0 0052	0 902[c]	1 055[g] ±0 081	0 167[c]	0 233[g] ±0 020	2 12[c]	
$_{82}$Pb	0 0537[a]	0 0508 ±0 0015	1 032[a]	1 052 ±0.015	0 228[a]	0 230 ±0 007	2 31[a]	2 34 ±0 03
	0 0537[d]	0 0513[g] ±0.0042	1 211[d]	1 203[g] ±0 104	0 284[d]	0 235[g] ±0 022	2 55[d]	
	0 0537[e]		1.114[e]		0 257[e]		2 43[e]	
	0 0537[f]		1 087[f]		0 251[f]		2 39[f]	

Table-5 5 Comparison of experimental L subshell ionization cross-sections (in barns) of Ta, Au and Pb with theoretical predictions of Scofield[9]

Target	σ_{L_1}		σ_{L_2}		σ_{L_3}		σ_{L_T}	
Atom(Z)	Theory	Expt	Theory	Expt	Theory	Expt	Theory	Expt
$_{73}Ta$	344 0	399 2[a]	170	270 4	197 2	264 5	711 2	934
		±12		±8		±8		±16
$_{79}$Au	436 3	552 6[a]	266 5	275 0	292 5	363 4	995 3	1191
		±15		±8		±10		±20
		438 0[b]		272 9		411 5		1122
		±12		±8		±12		±19
		778 1[c]		273 9		131 3		1183
		±20		±8		±5		±22
$_{82}$Pb	485 8	594 7[a]	328 9	410 9	351 0	414 2	1165 7	1419 8
		±18		±12		±12		±25
		493 4[d]		385 0		562.4		1440 8
		±15		±12		±17		±25
		493 4[e]		424 5		533 6		1451 4
		±15		±12		±16		±25
		459 5[f]		422 6		504 3		1386 4
		±15		±12		±15		±25

References a, b, c, d, e, f and g are same as in table-3

Table-5 6 Measured fluorescence parameters of Ta, Au and Pb

Target Element	ω_1	ω_2	ω_3	$\bar{\omega}_L$
$_{73}Ta$	0 169	0 392	0 332	0 336
$_{79}$Au	0 137	0 410	0 387	0 399
$_{82}$Pb	0 114	0 440	0 433	0 452

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