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The fluorescence, Auger, and Coster Kronig L_{II} subshell yields of thorium†

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Abstract. Fluorescence (ω_2), Auger (a_2) and Coster Kronig (f_{23}) L_{II} subshell yields in thorium have been determined from measurement on LX-rays emitted following α decay of ^{232}U. α–X coincidences techniques, employing a surface barrier detector for the α particles and NaI(Tl) scintillation counter for LX-rays, were used to measure the total fluorescence yield $\bar{\omega}_L$ of the L shell in ^{228}Th. The relative intensity of the LX photons from the L_{II} and the L_{III} subshells was determined with a Si(Li) LX-ray spectrometer. The experimental results are: $\omega_2 = 0{\cdot}44 \pm 0{\cdot}03$; $f_{23} = 0{\cdot}13 \pm 0{\cdot}10$; $a_2 = 0{\cdot}43 \pm 0{\cdot}10$, and $\bar{\omega}_L = 0{\cdot}46 \pm 0{\cdot}03$.

1. Introduction

Fluorescence, Auger, and Coster Kronig yields are important quantities used in nuclear spectroscopy and atomic physics, mainly in processes which involve the interaction of the nucleus with the orbital electrons, such as electron capture and internal conversion.

Burhop (1955), Listengarten (1960) and more recently Fink *et al.* (1966) published review articles, where the results of measurements of fluorescence yields have been summarized. The review by Fink *et al.* has shown that the different measurements of the L-shell yields do not agree well with each other. This lack of agreement is particularly relevant for the L_{II} subshell yields, mainly on the heavy elements. In the present work, experimental values of the L_{II} subshell and the total fluorescence yield $\bar{\omega}_L$ are reported; measurements have been undertaken on the LX-rays emitted following the α decay of ^{232}U. Halley and Engelkemeir (1964) measured previously the L shell fluorescence yield of thorium and the value obtained $\bar{\omega}_L = 0{\cdot}488 \pm 0{\cdot}008$, is not in agreement with the value 0·40, calculated by Kinsey (1948) using the widths of the level.

Knowledge of the decay scheme of ^{232}U (Lederer *et al.* 1967) and the relative intensities of the two strongest α particle groups emitted in the decay of this nucleus enables us to write the expression (1), which was used in the determination of $\bar{\omega}_L$

$$\bar{\omega}_L = \frac{N_\nu}{(0{\cdot}314 \pm 0{\cdot}004)N_\alpha(\Omega_x/4\pi)} \frac{1+\alpha}{\alpha_L} \frac{1}{t_x}. \tag{1}$$

N_α is the total α counting rate and N_ν the true coincidence counting rate of α particles and LX-ray photons, emitted by ^{232}U in the solid angle $\Omega_x/4\pi$. α and α_L are respectively the total internal conversion coefficient and the L internal conversion coefficient of the 57·9 keV E_2 transition from the first excited state to the ground state in ^{228}Th; the effect of the internal conversion of the very weak γ transitions of ^{228}Th is not significant, on the measurement of $\bar{\omega}_L$ and, therefore, has been neglected; t_x represents the LX-rays transmission coefficient through the source holder and the window of

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the crystal; $0{\cdot}314 \pm 0{\cdot}004$ is the relative intensity of the group of α particles which populate the 57·9 keV excited state of ^{228}Th and has been determined by us. The measurement of the fluorescence yields is simplified in this case, because, as was pointed out by Salgueiro *et al.* (1961), the primary ionization in a heavy even–even nucleus is effectively confined to the L_{II} and L_{III} subshells. The equations governing the L_{II} subshell fluorescence yields have been given by Salgueiro *et al.* in order to measure the yields of Pu. The ratio of the number of ionization vacancies in the L_{III} subshell to the number in the L_{II} subshell is represented by C_3' and the ratio of LX-ray photons from the L_{III} subshell to the number from the L_{II} subshell by F_3'. In terms of these quantities the relations of Salgueiro *et al.* may be written

$$\omega_2 = \bar{\omega}_L \frac{1+C_3'}{F_3'} \tag{2}$$

$$f_{23} = \omega_2 \frac{F_3'}{\omega_3} - C_3' \tag{3}$$

where ω_2, ω_3 and f_{23} denote respectively the fluorescence yields of the L_{II} and L_{III} subshells and the Coster Kronig yield for ionization transfer between these levels.

The Auger yield of the L_{II} subshell, a_2, has been calculated by

$$a_2 = 1 - \omega_2 - f_{23}. \tag{4}$$

In order to evaluate ω_2 and f_{23}, it is necessary to know the experimental quantities $\bar{\omega}_L$, C_3', F_3' and ω_3. Two of these, $\bar{\omega}_L$ and F_3', were measured by us; C_3' and ω_3 were estimated from previous work.

2. Experimental procedure

2.1. *Preparation of ^{232}U source*

The source of ^{232}U was prepared, by evaporation under vacuum and deposition on aluminium foils (5 μm thick), from a hydrochloric solution of this isotope obtained from the Radiochemical Centre (Amersham).

The difference between the boiling points of uranium and thorium enabled us to prepare sources where only ^{232}U and ^{224}Ra and its daughter products were present. Coincidence measurements were carried out on one of these sources two months after it had been prepared, and so only ^{232}U and a small fraction of ^{228}Th and its daughter products, which have been accumulated during this period, were present (figure 1).

The proportions of thorium and radium present in the ^{232}U peak were determined from the relative intensity of the ^{224}Ra and ^{228}Th α peaks, emitted by a ^{228}Th source in equilibrium with its daughter products (figure 2); ten different spectra were used and this relative intensity was found to be $0{\cdot}88 \pm 0{\cdot}02$, in good agreement with the value which comes from the radioactive equilibrium equations. In the α spectrum of the ^{232}U source, the (^{232}U + ^{228}Th) pulses are well resolved from the ^{224}Ra strongest pulse (94·5%); so, by means of the previous value $0{\cdot}88 \pm 0{\cdot}02$ and the knowledge of the counting rate of this ^{224}Ra pulse, it has been possible to deduce, from the total α counting rate, the fraction which is due to ^{228}Th and to the α group (5·5%) which populates the first excited state of ^{224}Ra.

2.2. *Experimental arrangement*

2.2.1. *Measurement of* $\bar{\omega}_L$. The α particles were detected in an Ortec surface barrier detector, and LX-rays were recorded in a scintillation crystal Harshaw type HG, through a beryllium window 0·127 mm thick and 1 μm of bright aluminium foil reflector; a photomultiplier EMI 9514 S was used. The source and detectors were

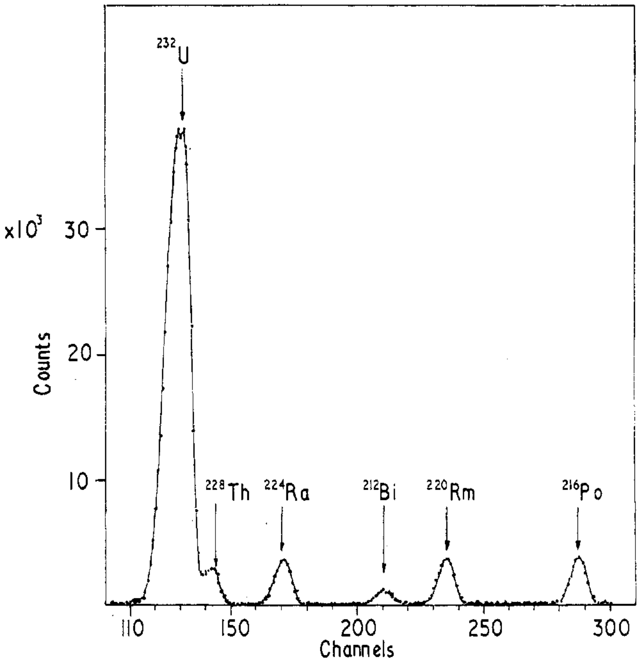

Figure 1. Spectrum of ^{232}U source used in coincidence experiments.

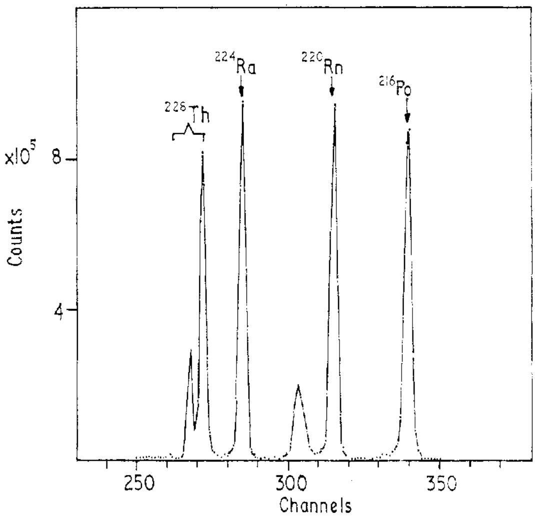

Figure 2. Spectrum of a ^{228}Th source in equilibrium with its daughter products.

placed inside a vacuum chamber in such a way that they all had the same symmetry axis. Special arrangements were used to change and determine carefully the relative positions of the detectors and the source so that the geometrical detection conditions could be defined very accurately. In a previous investigation (Gil *et al.* 1965) the limit of proportionality between the photon counting rate and the value of the solid angle was found to be $\Omega_x/4\pi = 0{\cdot}0106$; in this research the solid angles were always less than this value. Corrections, due to the finite size of the source could be neglected in view of the source–crystal distances which have been used. It has been shown by Falk-Vairant *et al.* (1954) and Halley and Engelkemeir (1964) that the angular correlation of α particles and LX-rays from the decay of a heavy even–even nucleus is isotropic within the experimental errors (4%). We assume therefore that in our experiments also the α–X angular correlation of ^{232}U is isotropic. A slow conventional coincidence spectrometer composed of Ortec units has been employed.

2.2.2. *Measurement of F_3'.* F_3' was determined using a Si(Li) detector (Ortec model 7014–06315) having 6 mm of active diameter, a sensitive depth of 3 mm and a window of beryllium 0·13 mm thick. The detector was placed outside the vacuum chamber, facing a 9·55 mg cm^{-2} mica window, which was sealed on the wall of the chamber; cylindrical symmetry was maintained between the detector and the source.

The LX-ray photons emitted by ^{228}Th have energies in the range 10 to 16 keV. Attenuation corrections, for different absorption of the LX photons in the windows of beryllium and mica and in the air, have been made. The efficiency of the Si(Li) detector, in the range 10–20 keV, was found to be constant within the experimental errors, in good agreement with previous results presented by different authors (Freund *et al.* 1969, Hollstein 1970).

3. Experimental results

3.1. *Value of $\bar{\omega}_L$*

A weighted mean of the results of 14 coincidence experiments enabled us to determine $N_v/N_\alpha(\Omega_x/4\pi) = 0{\cdot}102 \pm 0{\cdot}004$. This value, corrected for the photon absorption in the source holder and in the window of the detector, became $0{\cdot}105 \pm 0{\cdot}004$. From the results of Halley and Engelkemeir (1964) and Duke and Talbert (1968) one can easily deduce that $\alpha_L/(1+\alpha) = 0{\cdot}726 \pm 0{\cdot}046$. Therefore expression (1) gives $\bar{\omega}_L = 0{\cdot}46 \pm 0{\cdot}03$. The relatively large error (more than 6%) in the present determination of $\bar{\omega}_L$ is mainly due to uncertainties in the values of internal conversion coefficient; in fact, an error of less than 4% was attributable to our coincidence-counting measurements.

Although the present value for $\bar{\omega}_L$ is slightly less than Halley's result there is agreement within the experimental errors. However, in the work of Halley *et al.* it is possible that the limit of proportionality between the photon counting rate and the solid angles may have been exceeded.

3.2. *Values of ω_2, f_{23} and a_2*

The fluorescence yield ω_2 of the L_{II} subshell was determined from expression (2), where C_3' was estimated from previous work, summarized in table 1; the value $C_3' = 0{\cdot}862 \pm 0{\cdot}030$ has been adopted. The table shows conclusively that the L_I conversion of 57·9 keV γ radiation may be neglected within the experimental errors.

The LX-ray spectrum in coincidence with α particles of ^{232}U is shown in figure 3. The L_l, L_α, L_β and L_γ X-ray groups are clearly resolved in the spectrum presented

Table 1

	L_I	L_{II}	L_{III}	References
Experimental value	$0{\cdot}0355 \pm 0{\cdot}0025$	$1{\cdot}15 \pm 0{\cdot}04$	1	Hamilton *et al.* 1966
Theoretical values	$0{\cdot}038$	$1{\cdot}18$	1	Nuclear data 1966
	$0{\cdot}0372$	$1{\cdot}15$	1	Hamilton *et al.* 1966
Theoretical mean values	$0{\cdot}0376$	$1{\cdot}16$	1	
Adopted values	$0{\cdot}0365 \pm 0{\cdot}026$	$1{\cdot}16 \pm 0{\cdot}04$	1	

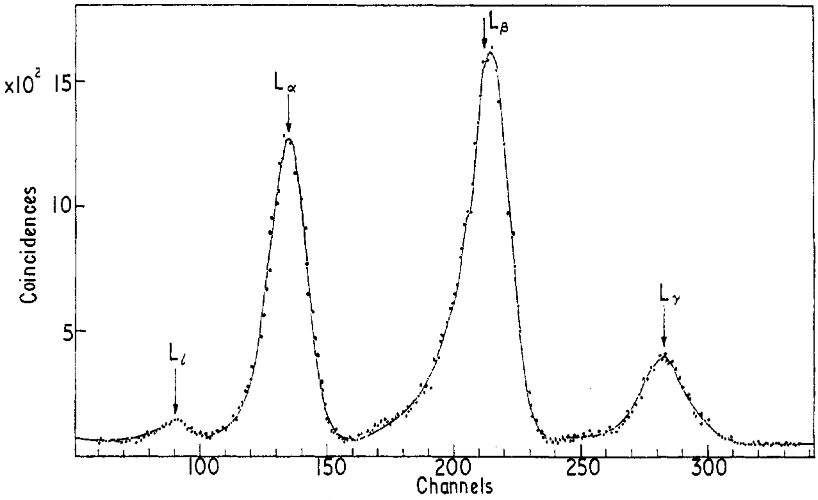

Figure 3. α–X coincidences spectrum of ^{232}U.

in figure 3. L_l and L_α groups $(L_{\alpha_1}+L_{\alpha_2})$ are due to primary ionizations in the L_{III} subshell. The principal lines which belong to the L_β group are due to L_{II} vacancies (mainly L_{β_1}) and L_{III} primary ionizations $(L_{\beta_2}+L_{\beta_{15}}, L_{\beta_5}, L_{\beta_6})$. This last component of the L_β group will be designed further by $L_{\beta'}$. In order to find a value of F_3' we must know the number of LX-ray photons from the L_{III} subshell $F(L_{III})$ and the number from the L_{II} subshell $F(L_{II})$, in ^{228}Th; this could be done, in our experiment, by deducting from the L_β group the contribution due to $L_{\beta'}$, which can be evaluated from the results of Compton and Allison (1946) and Goldberg (1962). In fact, according to these authors, the ratio of the number of X-ray photons in the $L_{\beta'}$ group, $F(L_{\beta'})$, to the number in the L_α group, $F(L_\alpha)$, is $0{\cdot}33 \pm 0{\cdot}03$. $F(L_{\beta'})$ could then be calculated from the experimental value of $F(L_\alpha)$ measured in the coincidence spectrum (figure 3). Therefore, as $F(L_{\beta_1}) = F(L_\beta) - F(L_{\beta'})$ we can write finally $F(L_{II}) = F(L_{\beta_1}) + F(L_\gamma)$ and $F(L_{III}) = F(L_l) + F(L_\alpha) + F(L_{\beta'})$.

So we found $F_3' = 0{\cdot}95 \pm 0{\cdot}04$, which is a mean value of three different determinations. Substituting in equation (2) the values of $\bar{\omega}_L$, F_3' measured in this work and C_3' adopted by us, we obtain $\omega_2 = 0{\cdot}44 \pm 0{\cdot}03$. The following results of f_{23} and a_2 are found from equations (3) and (4) using the value $\omega_3 = 0{\cdot}42 \pm 0{\cdot}02$ determined by Stephenson (1937) and Haynes and Achor (1955):

$$f_{23} = 0{\cdot}13 \pm 0{\cdot}10$$

$$a_2 = 0{\cdot}43 \pm 0{\cdot}10.$$

4. Conclusions

The present result for the fluorescence yield ω_2 is in close agreement with the value obtained by Price *et al.* (1968), but both results are smaller than the calculated values. Theoretical values for ω_2 predicted by Listengarten (1960) are usually much larger than the experimental results with the exception of the ω_2 value measured by Byrne *et al.* (1968). Listengarten assumes that f_{23} is zero for thorium, due to the fact that $L_{II} \to L_{III}\, M_{IV,V}$ transitions are energetically possible only for $Z \geqslant 91$. The f_{23} value obtained in the present work shows undoubtedly that f_{23} is not zero for thorium and this is the significant result as far as f_{23} is concerned. Experimental values for f_{23} are usually much higher than theoretical ones, although Salgueiro's result for plutonium agrees with the calculated value. The Auger yield a_2 obtained in the present work is in particularly good agreement with the theoretical value (Listengarten 1960).

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