L X-ray Spectra and E2 Internal Conversion Coefficients in Radon and Radium

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(Z. Naturforsch. **28 a**, 1635–1641 [1973]; received 9 April 1973)

The X-ray spectra resulting from the internal conversion of electric quadrupole transitions following the alpha decay of Th^{230} and Ra^{226} were analysed with a Si(Li) spectrometer. From the knowledge of the Coster-Kronig and fluorescence yields, the internal conversion coefficients of the E2 transitions from the first excited states in Ra^{226} and Rn^{222} could be deduced. Results are in good agreement with theoretical values.

I. Introduction

When a vacancy is produced in one of the atomic L-subshells, there are three processes the atom can undergo in transferring it to an outer orbit. First, an electron from a higher shell can fill the L_i-subshell vacancy in a radiative transition, resulting in an L_i X-ray. The probability of this process is measured by the fluorescence yield ω_i. Second, the vacancy may be filled by an electron from a higher shell, the difference in binding energy being utilized in the ejection of an outer electron from the atom. Such an ejected electron is known as an Auger electron and the probability of this process is measured by the Auger yield a_i. Third, the vacancy may be filled by an electron from a higher L-subshell, the energy being used to eject an electron from an outer shell. This process is known as a Coster-Kronig transition and its probability is measured by the CK yield $f_{ij} (j>i)$. Obviously $\omega_i + a_i + \sum_j f_{ij} = 1$, for $i, j = 1, 2, 3$.

Let us consider a nuclear transition with such energy as can be internally converted only in the L- (or higher) subshells. If the internal conversion coefficients in the L subshells, α_i, are known, we can calculate the number of primary vacancies produced in each of the L subshells. This number, N_i, is the product $\alpha_i N_\gamma$ where N_γ is the intensity of the unconverted gamma-ray. If, on the other hand, it is possible to measure the intensities of the X-rays, i.e., the quantities, $N(L_i)$, which represent the number of X-rays resulting from the radiative filling of a vacancy in L_i, then informations can be obtained about the radiative and non-radiative yields.

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Conversely, if the $N(L_i)$ and N_γ intensities are measured and all the fluorescence and Auger yields are known, then the internal conversion coefficients (ICC) in each L-subshell can be determined provided the L-X-rays are due to a single nuclear transition.

The resulting average fluorescence yield is the ratio

$$\bar{\omega}_L = X_L/\alpha_L = \sum_i X_i / \sum_i \alpha_i \quad \text{were} \quad X_i = N(L_i)/N_\gamma .$$

If the internal conversion in the K-shell is also energetically possible then the filling of the resulting K-vacancies can create vacancies in the L-subshells through the emission of K_α X-rays or K Auger electrons. It should be noted that an L electron filling a K-vacancy could give rise to an Auger electron from a higher L-subshell (a K-LL Auger electron) which would leave the atom with two L-shell vacancies.

In this case the ICC's in the L-subshells can be determined from the knowledge of the nine quantities X_i, ω_i and f_{ij} more the new quantities X_K, ω_K and f_{Ki} related to the K-shell. Following the same notation as before, $X_K = N(K)/N_\gamma$ and $\omega_K = X_K/\alpha_K$ is the K-shell fluorescence yield, α_K being the K-shell ICC.

The quantity f_{Ki} is the number of L_i vacancies created by one primary K-vacancy. If we define the K-Auger yield $a_K = 1 - \omega_K$ then:

$$f_{Ki} = [\omega_{Ki} + a_K (K\,L_i\,X + K\,X\,L_i)/\text{K-Augers}] + 2\,a_K\,K\,L_i\,L_i/\text{K-Augers}$$

where ω_{Ki} is the partial fluorescence yield given by ω_K times the ratio of $K L_i$ X-rays to all K X-rays. The intensities of the Auger electrons ejected from Z when a vacancy is shifted from X to Y is represented by XYZ. In the above expression, $K L_i X$ means the sum over all the energetically possible X

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orbits ($\neq L_i$) and "K-Augers" is the total intensity of the Auger electrons.

The average fluorescence yield $\bar{\omega}_L$ is now given by

$$\bar{\omega}_L = X_L/(\alpha_L + \alpha_K f_K) \quad \text{where} \quad f_K = \sum_i f_{Ki}.$$

This method allows the determination of the absolute values of the ICC's by a simple analysis of the singles spectrum covering the L X-rays (eventually the K X-rays too) and the single unconverted gamma-ray. However it implies the knowledge of the six quantities ω_i and f_{ij} (eventually the quantities ω_K and f_{Ki} too).

Even when the unconverted gamma-ray is not observed it is possible to find the ratios $\alpha_1/\alpha_2/\alpha_3$ and, if a reasonable value $\bar{\omega}_L$ is adopted, we can calculate α_L and then estimate the subshell ICC's. If the nuclear transition is unknown we can, on the basis of the ICC's, determine the multipolarity and even the energy of the transition. This method requires high resolution X-rays detectors in order to allow the identification of the individual lines we need to compute the $X(L_i)$ intensities. For high Z atoms these lines appear in three main groups known as L_α, L_β and L_γ groups. The L_α group is due to radiative transitions to the L_3 subshell, the L_γ group is composed by radiative transitions to L_1 and L_2 subshells and the L_β group, by far the more complex, is related to the three subshells.

Even when only the most prominent lines can be isolated and measured, the situation is not hopeless. If, at least, one radiative transition to each subshell is identified theoretical branching ratios [1, 2] can be used to estimate the $X(L_i)$ intensities. Recent experimental works [3–6] show that these theoretical branching ratios are quite reliable (see comments in Section III – c).

A similar method, based on the $L_\alpha/L_\beta/L_\gamma$ ratios, was suggested by Clark and Stabenau [7, 8] and applied to the 0,28 sec isomeric transition in Ta^{182}. The resolution of the spectrometer used by them was not good enough to allow the identification of the individual L X-rays lines but the α, β, and γ groups were well separated. They adopted the branching ratios tabulated by Storm and Israel [9], the conversion coefficients interpolated from the theoretical values of Hager and Seltzer [10] and fluorescence and CK yields given in the literature. The L X-ray intensity ratios β/γ and γ/α were then calculated for Tantalum and ploted as a function of the transition energy, for each multipolarity. In some favorable conditions it seems to be possible to determine the multipolarity and energy of low energy (bellow K-shell binding energy), highly converted nuclear transitions for which the unconverted gamma-ray are not observed.

II. Experimental Procedures

In this paper we report the determination of the internal conversion coefficients of two electric quadrupole transitions: 67,68 KeV in Ra^{226} and 186,5 KeV in Rn^{222}. These transitions are from the first excited 2^+ state to the ground state. The levels in Ra^{226} and Rn^{222} were populated by the α decay of Th^{230} and Ra^{226}, respectively.

Thin carrier-free sources were prepared from commercially available (Radio Chemical Centre, Amersham, UK) sources of Th^{230} and Ra^{226}, after removing the descendants. The contamination of Th^{232} in the source of Th^{230} was estimated in 5%. Absorption in the source was supposed to be negligible as well as the production of K or L vacancies by alpha particles ionization.

Possible vacancies produced by the internal conversion of nuclear transitions other than the two aforementioned were not considered. The error introduced by this approximation in the ICC's is probably less than 1%.

The singles spectra were studied with an ORTEC Si(Li) spectrometer which has a resolution of 180 eV full width at half maximum (FWHM) for 6.4 KeV Fe K_α X-rays from Co^{57}. The detector has a sensitive depth of 3 mm and an active diameter of 4 mm and is enclosed in a housing with a 0.025 mm Be window and a 200 Å gold contact. The photopeak relative efficiency curve of the detector was obtained in the usual way with standard radioactive sources presenting low energy transitions with well known intensities. The efficiency curve covers a region from 3 to 140 KeV. The spectrometer was observed to have an almost flat response for photopeak detection in the energy range from 8 to 22 KeV, and so was ideally suited for measurements of relative L X-ray intensities of heavy elements.

We also employed an ORTEC Ge(Li) spectrometer with a Beryllium window 0.25 mm thick and a resolution of 700 eV FWHM for the 122 KeV gamma-rays of Co^{57}. The $N(L)/N(K)$ ratio in Rn^{222} was found to be 1.173 ± 0.044 with the first spectrometer and 1.195 ± 0.036 with the second one.

Most of the individual L_α, L_β and L_γ lines were not fully resolved, and hence a peak fitting procedure had to be used to extract accurate values for the intensities of the various X-ray lines. The energies adopted[11] for the L X-rays are given in Tables 3 and 5. A graphical peeling method was used and full-energy-peak profiles were determined experimentally for different sections of the spectra. The FWHM was observed to vary linearly with the energy in the small interval from 10 to 22 KeV and was determined by interpolation for each value of the energy of a particular L X-ray line. The low energy tail of the profiles was carefully determined for each interval of 3 KeV in the same energy range. The relative intensities of the most prominent lines could be determined with very small relative errors (3 to 4%). For weaker lines, however, these errors can be as high as 10 or even 20%. Since the weight of these last lines is small in the computation of the integrated $N(X_i)$ intensities the errors in the number of L_i X-rays could be reduced to 4 to 5% (except for $i = 1$) with a resulting error of 5 to 7% in the X_i ratios.

In calculating the $N(X_i)$ intensities use was made of the theoretical branching ratios of Scofield[1] in order to consider the weakest transitions which can not be experimentally observed due to poor statistics and/or resolution.

Values for the L shell fluorescence yields and CK transition probabilities are poorly known in general. However, reliable experimental values for some heavy elements were recently published by several authors[6, 12–23]. Theoretical calculations[24–27] can also be used as a guide. For an E2 transition, since α_1 is expected to be small as compared with α_2 and α_3, the most relevant yields are ω_2, ω_3 and f_{23}. Fortunately the L_2- and L_3-subshell fluorescence yields and the $L_2 - L_3 X$ Coster-Kronig transition probability are by for the best known in the high-Z atoms. The adopted ω_i and f_{ij} employed in this work are given in Table 1 (see also Figs. 1 to 3). Theoretically the fluorescence yields ω_2 is expected to drop sharply at $Z \cong 91$, where transitions of the $L_2 - L_3 M_5$ type become energetically possible, and

Table 1. Adopted values of the fluorescence and Coster-Kronig yields in Rn and Ra.

Yield	$Z = 86$ (Radon)	$Z = 88$ (Radium)
ω_1	0.15	0.16
ω_2	0.41	0.43
ω_3	0.40	0.42
f_{12}	0.10	0.10
f_{13}	0.65	0.66
f_{23}	0.14	0.14

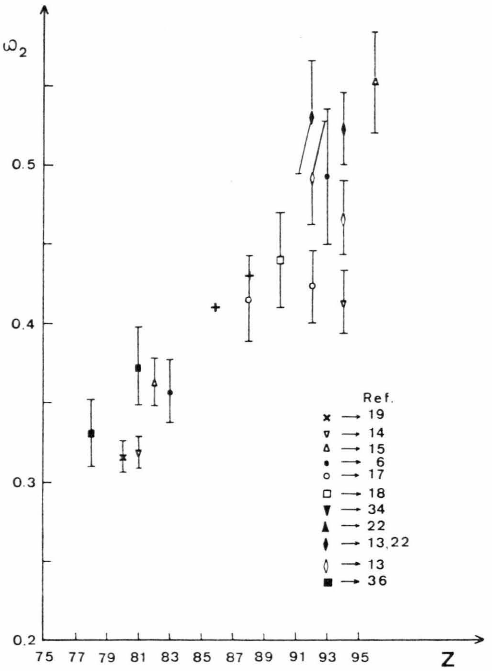

Fig. 1. L_2-subshell fluorescence yield ω_2 as a function of atomic number. The symbol + refers to the adopted values for Rn and Ra.

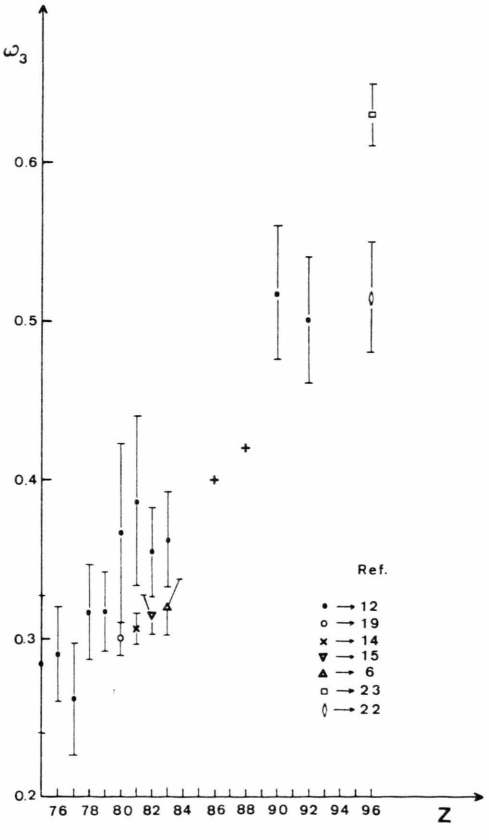

Fig. 2. L_3-subshell fluorescence yield ω_3 as a function of atomic number. The symbol + refers to the adopted values for Rn and Ra.

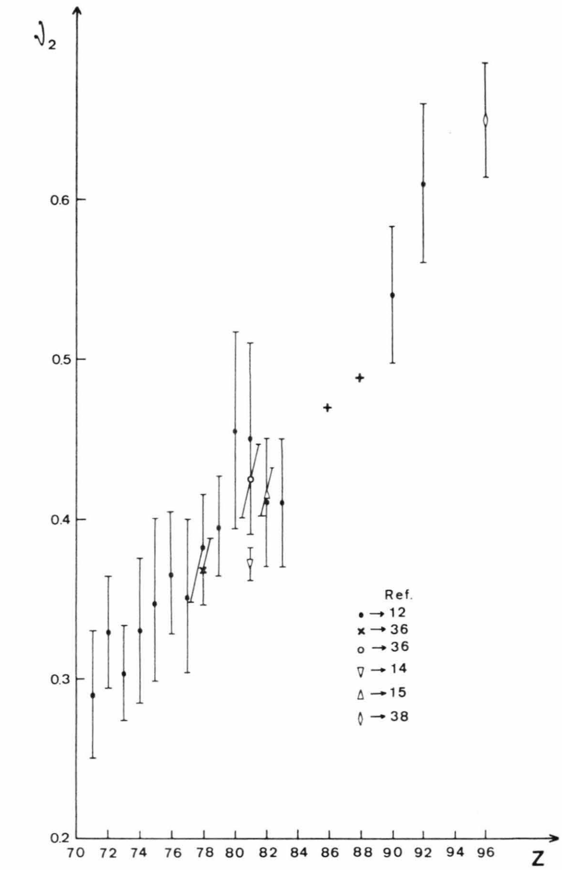

Fig. 3. X-ray $\nu_2 = \omega_2 + f_{23}\,\omega_3$, as a function of atomic number. The symbol + refers to the adopted values for Rn and Ra.

consequently an abrupt discontinuity must be present in the f_{23} vs. Z curve. Experimentally this is not confirmed and only smooth changes in the slope of the curves of ω_2 and f_{23} vs. Z are observed.

There is a direct measurement of the L_2-subshell yields in Ra by Gil et al. [17]. The L X-rays following the α decay of Th^{228} were analysed by those authors with the following results: $\omega_2 = 0.415 \pm 0.027$ and $f_{23} = 0.01 \pm 0.07$. Essentially they observed the L X-rays due to the internal conversion of the 84.4 KeV E2 transition in Ra^{224}. The measured value of ω_2 is in close agreement with the adopted one, but f_{23} is too small even though a large experimental error is assigned.

The K fluorescence yield is relatively well determined through the entire periodic table. In the high Z region it seems to be nearly constant and equal to 0.97. We choose the value $\omega_K = 0.973$. In a previous work [28] we measured the $K\alpha_2/K\alpha_1$ and K_β/K_α ratios in Rn. We repeated the experiment obtaining essentially the same results, namely: $K\alpha_2/K\alpha_1 = 0.594$ and $K_\beta/K_\alpha = 0.287$. From this we get $\omega_{K1} = 0$; $\omega_{K2} = 0.282$ and $\omega_{K3} = 0.474$.

The L vacancies following K-Auger electron emission are the least well known of all the quantities involved in this experiment. A critical summary of the available K-Auger electron relative intensities has been given by Bergstrom and Nordling [29]. In spite of the lack of precise knowledge of the Auger electron intensities, contribution to the error in the present measurements is small, since fewer than 3% of the K-shell vacancies are filled by Auger transitions. We define

$$a_{KXY} = a_K \cdot KXY/K\text{-Augers}\,. \qquad (7)$$

The adopted values of a_{KXY} are given in Table 2. Finally we get $f_{K1} = 0.017$, $f_{K2} = 0.294$, $f_{K3} = 0.799$. We suppose f_K to be correct within 3%.

Table 2. Adopted values of the partial Auger yields in Rn.

XYZ	a_{xyz}
K L_1L_1	0.0030
K L_1L_2	0.0045
K L_1L_3	0.0029
K L_2L_2	0.0008
K L_2L_3	0.0040
K L_3L_3	0.0014
K L_1S (*)	0.0032
K L_2S (*)	0.0024
K L_3S (*)	0.0037
K S T (*)	0.0015

(*) S, T ≠ L.

III. Results and Discussion

a) Radium

The measured intensities of the L X-ray lines of Ra are presented in Table 3. They are arbitrarily normalized making the intensity of the β_1 line equal to 100. The L X-ray spectrum is shown in Figure 4.

The $L_l + L_\alpha/L_\eta + L_\beta/L_\gamma$ ratios were found to be $76.4 \pm 2.9/100/22.5 \pm 1.2$.

In Table 4 we present the relative intensities X_i and the resulting values of a_i. The agreement with the theoretical values [10] is quite satisfactory.

With the Th^{230} source the X_L/X_K ratio was found to be 310 ± 20 thus justifying the neglecting of K vacancies in the computation of the L_i-internal conversion coefficients. Some results about the internal conversion of the 67.68 KeV E2 transition in Ra^{226} have been previously published. Rosenblum et al. [30] report the ratio $L_2/L_3 = 1.0$ and Rester et al. [31] give

Table 3. Relative intensities of Ra L X-rays arising from the α-decay of Th^{230}.

Line	Energy (KeV)	Relative Intensity
L_3M_1	10.622	4.95 ± 0.19
$L_3M_{4,5}$	12.330	92.95 ± 2.95
L_2M_1	13.663	2.60 ± 0.16
L_3N_1	14.235	1.33 ± 0.09
L_1M_2	14.415	0.65 ± 0.05
$L_3N_{4,5}$	14.82	18.90 ± 1.00
L_2M_4	15.236	100.00 ± 2.00
$L_3O_{4,5}$	15.38	3.30 ± 0.22
L_1M_3	15.444	0.7 ± 0.1
L_2N_1	17.275	0.8 ± 0.1
L_2N_4	17.848	23.31 ± 0.98
L_1N_2, L_2N_6, L_2O_1	18.20	0.7 ± 0.1
L_2O_4	18.417	4.02 ± 0.18
$L_1O_{4,5}$, L_1P	19.17	0.13 ± 0.02

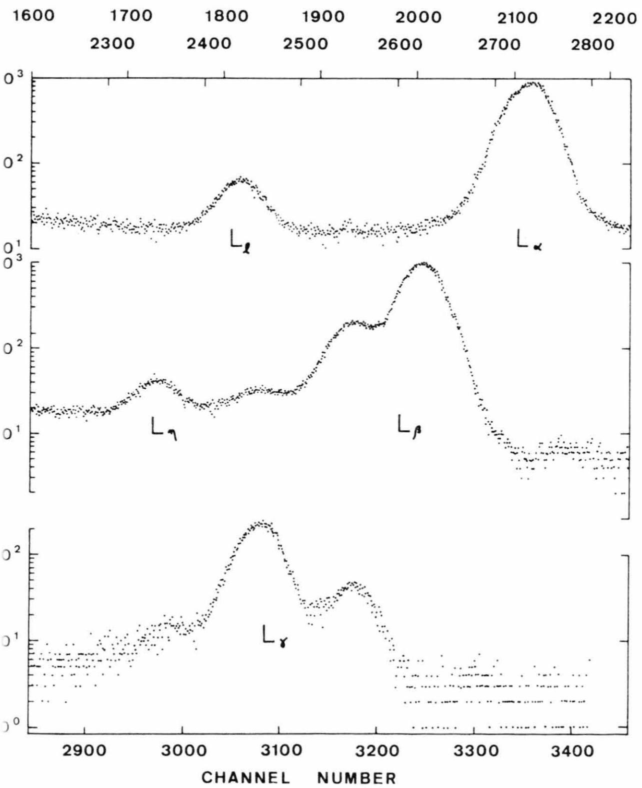

Fig. 4. Radium L X-ray spectrum associated with the α-decay of Th^{230}. This spectrum corresponds to a run of 62 hours, after the subtraction of the background.

$(L_1+L_2)/L_3=(13\pm 2)/10$. Our result is $L_1/L_2/L_3=0.47/12.5/10$. Booth et al. [32] had obtained $X_L=17.2$ and then $\alpha_L=46\pm 5$ with $\bar{\omega}_L=0.52\pm 0.05$.

Table 4. Number of L X-rays per unconverted gamma-ray and the resulting internal conversion coefficients of the $2^+ \rightarrow 0^+$ transition in Ra^{226}.

Radium Subshell	X_i	α_i Experimental	α_i Theoretical [10]
L_1	≃ 0.15	≃ 0.9	0.76
L_2	10.77 ± 0.70	25.0 ± 2.0	24.75
L_3	10.03 ± 0.37	19.8 ± 1.8	19.50
L (Total)	21.0 ± 0.8	45.6 ± 4.5	45.0

As a final result we get $\bar{\omega}_L=0.461\pm 0.054$. Since the measured value of α_L agrees well with the theoretical value we can reduce the error in $\bar{\omega}_L$ by taking the experimental X_L ratio and the theoretical value of α_L. Then, $\bar{\omega}_L=0.467\pm 0.024$.

Beside the value of Booth et al. [32] for $\bar{\omega}_L$ other experimental results are found in the literature (see Table 7).

b) Radon

The measured intensities of the L X-ray lines of Rn are presented in Table 5 with the same normalization employed in the Table 3. We note that now the contribution of the L_1 X-ray lines is, in average, 2.5 times more important than in Ra. The L X-ray spectrum is show in Figure 5.

Table 5. Relative intensities of Rn L X-rays arising from the α-decay of Ra^{226}.

Line	Energy (KeV)	Relative Intensity
L_3M_1	10.137	4.97 ± 0.18
$L_3M_{4,5}$	11.716	95.62 ± 2.95
L_2M_1	12.855	2.58 ± 0.16
L_3N_1	13.522	1.42 ± 0.10
L_1M_2	13.890	1.90 ± 0.12
$L_3N_{4,5}$	14.06	19.20 ± 1.15
L_2M_4	14.315	100.00 ± 2.00
L_1M_3	14.510	1.85 ± 0.13
$L_3O_{4,5}$	14.58	2.95 ± 0.19
$L_1M_{4,5}$	15.12	0.20 ± 0.05
L_2N_1	16.240	0.8 ± 0.2
L_2N_4	16.77	22.00 ± 1.05
L_2N_6, L_1N_2, L_2O_1	17.12	0.9 ± 0.2
L_1N_3, L_2O_4	17.30	4.05 ± 0.30
$L_1O_{4,5}$, L_1P	18.00, 18.04	0.30 ± 0.03
KL_2	81.067	61.92 ± 2.10
KL_3	83.786	104.30 ± 2.93
KM_2	94.245	12.15 ± 0.60
KM_3	94.866	23.26 ± 1.18
$KM_{4,5}$	95.45	0.76 ± 0.19
KN	97.6	9.02 ± 0.17
KO	98.3	1.80 ± 0.17

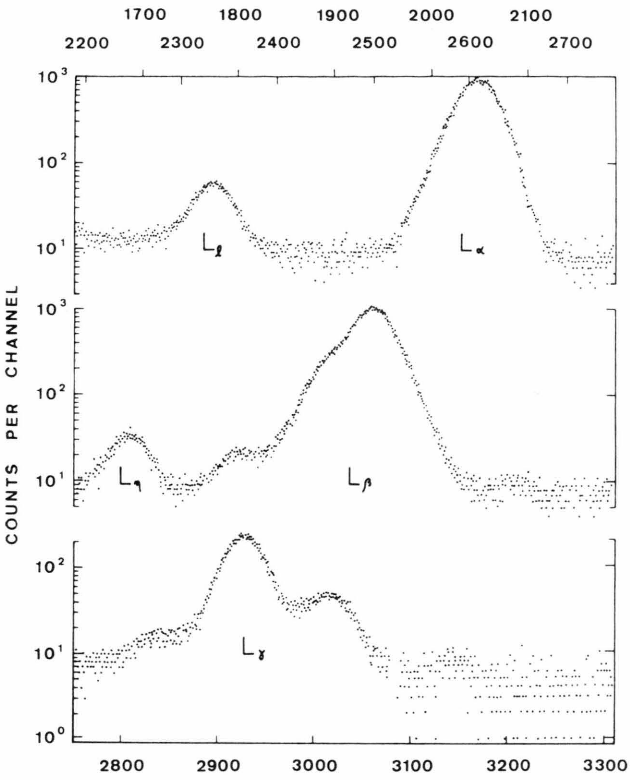

Fig. 5. Radon L X-ray spectrum associated with the α-decay of Ra^{226}. This spectrum corresponds to a run of 44 hours, after the subtraction of the background.

The ratios $L_l + L_a / L_\eta + L_\beta / L_\gamma$ are now $77.0 \pm 2.5 / 100 / 21.3 \pm 1.2$, then equal to the ratios found in Ra within the experimental errors.

In the Table 6 we present the relative intensities X_i and the resulting values of the internal conversion coefficients. Again, the agreement with the theoretical values [10] is rather good.

Table 6. Number of L X-rays per unconverted gamma-ray and resulting internal conversion coefficients of the $2^+ \to 0^+$ transition in Rn^{222}.

Radon Shell or Subshell	X_i	α_i Experimental	Theor.[10]
K	0.195 ± 0.007	0.200 ± 0.009	0.191
L_1	0.0051 ± 0.0007	0.031 ± 0.006	0.034
L_2	0.118 ± 0.008	0.226 ± 0.016	0.222
L_3	0.113 ± 0.004	0.124 ± 0.008	0.125
L (Total)	0.237 ± 0.012	0.380 ± 0.020	0.382

Gonçalves [33] had obtained for the 185.7 KeV E2 transition in Rn^{222} $K/L = 0.76$ and $\alpha_K = 0.22 \pm 0.02$. Our value of K/L is 0.526 ± 0.040, but good agreement is found for α_K.

For the average L-fluorescence yield we found $\bar{\omega}_L = 0.439 \pm 0.048$. Adopting the theoretical values of α_L and α_K and the measured values of the ratios X_L/N_γ, $X_{K\alpha}/N_\gamma$ and K_α/K_β we get $\bar{\omega}_L = 0.432 \pm 0.022$.

The number $\bar{\omega}_L$, of X-rays per L-shell vacancy is a quantity which depend on the mode of vacancy production. It is therefore different in internal conversion, e-capture, photo-processes or ionization by charged particles. In the internal conversion processes it depends on the multipolarity and energy of the nuclear transition. However, since measurements of $\bar{\omega}_L$ are rather scarce we present in Table 7 a survey of this quantity in the high-Z region when the L X-rays are due to the internal conversion of E2 transitions following the α decay of the parent nuclide.

Table 7. Average L-fluorescence yields from internal conversion of E2 transitions.

Element	Daughter Nucleus	ω_L	Ref.
Radon	Rn^{222}	0.432 ± 0.022	P.W.
Radium	Ra^{224}	0.404 ± 0.024	17
	Ra^{22r}	0.480 ± 0.012	35
		0.467 ± 0.024	P.W.
Thorium	Th^{228}	0.488 ± 0.008	35
		0.46 ± 0.03	18
Uranium	U^{234}	0.478 ± 0.009	35
		0.442 ± 0.012	17
		0.576 ± 0.015	13
	U^{23r}	0.570 ± 0.019	13
Plutonium	Pu^{238}	0.486 ± 0.01	34
	Pu^{240}	0.540 ± 0.009	35
		0.566 ± 0.010	13
Curium	Cm^{248}	0.531 ± 0.010	35

c) General Remarks

Close agreement between experimentally determined and theoretically calculated values of the internal conversion coefficients of pure E2 transitions is a general rule. In despite of the simple procedure employed in this paper to choose the relevant values of the fluorescence and Coster-Kronig yields and of the relatively large errors associated with the peeling method in the bad statistics portions of the spectra, the surprisingly good agreement we found for our measured values of α_i and the calculated values of Hager and Seltzer suggests that the adopted values of ω_2 and ω_3 are good within about 8%.

A suplementary remark concerns the measured branching ratios. The quantities s_i, defined by Rao et al.[20] represent, for each subshell L_i, the ratios of X-rays transitions originating from higher shells (N + O + ...) to transitions originating from M-subshells. As for the K_β/K_α ratio they seem to exceed Scofield's theoretical ratios[20, 38]. In the present case we found $s_2 = 0.268$ and $s_3 = 0.234$ for Rn and $s_2 = 0.279$ and $s_3 = 0.240$ for Ra. The s_3 values are not different from the theoretical results but the s_2 values are systematically (5 to 10%) greater than Scofields ratios, as observed by us (6) in Bi and Np (the errors in s_2 are of the same order of magnitude). The errors in s_i are too big to permit any conclusion.

Acknowledgments

It is a pleasure to acknowledge people from CNEN in both I.E.A. – SP and L.D. for helping us with sources and instrumentation. This work was performed under the auspices of the BNDE and CNPq.

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