

# CAPMULT, the counting efficiency for electron capture by a KLMN four-shell model

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## Abstract

We present a new more advanced program to compute the liquid scintillation counting efficiency for pure electron capture nuclides. We consider a KLMN four-shell model with 262 different ways of atomic rearrangement, which permits one to improve the accuracy of the method for nuclides of large atomic numbers.

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## PROGRAM SUMMARY

*Title of the program:* CAPMULT

*Catalogue number:* ADBH

*Program obtainable from:* CPC Program Library, Queen's University of Belfast, N. Ireland (see application form in this issue)

*Licensing provisions:* none

*Computer:* any IBM compatible with 80386 32-bit processor

*Operating system:* MS-DOS 3.30 and higher

*Programming language used:* FORTRAN 77

*Memory required to execute with typical data:* 22 kwords

*No. of bits in a word:* 32

*No. of lines in distributed program, including test data, etc.:* 1226

*Keywords:* Electron capture; Counting efficiency; Liquid scintillation; KLMN-shell model

### *Nature of the physical problem*

The computation of the liquid scintillation counting efficiency for electron capture nuclides (EC) requires the determination of the atomic pathways that follow the capture of one electron by the nucleus. The large number of different ways of atomic rearrangement in the daughter nuclide makes necessary the use of shell models in which subshells are averaged. A KLM three-shell model involves only 22 pathways, but it is inaccurate for nuclides of high atomic numbers. We propose a more advanced model with 262 pathways in which N-shell is included.

### *Method of solution*

The determination of the probabilities and effective energies for all pathways requires the prior evaluation of their analytical expressions. Since the number of expressions is large for a KLMN-model, we achieve all these expressions by using a computer program.

### *Restrictions of the complexity of the problem*

We average L, M and N subshells. We do not consider the atomic rearrangement for N and outer shells.

### *Typical running time*

The test run requires about 4 minutes on an IBM AT.

## LONG WRITE-UP

### 1. Introduction

The determination of the counting efficiency for electron capture (EC) and internal conversion (IC) by a simple KLM three-shell model [1–2] is in excellent agreement with experiment for low atomic number nuclides; i.e.  $^{55}\text{Fe}$ ,  $^{65}\text{Zn}$  or  $^{51}\text{Cr}$  [3]. However, such a model is insufficient for large  $Z$ -values. One important limitation arises from the enhancement of X-ray energies for large atomic numbers. The photoelectric interaction probability drops down and becomes very much dependent on the chemical components of the scintillator [4–5]. The use of Monte Carlo simulations permits one to compute the energy distribution of photoelectric and Compton electrons for these high energy X-rays [6]. On the other hand, since the fluorescence yield for L- and M-shells is low, the electron capture process for these shells generates mainly low energy Auger electrons, which are within a wide energy interval for large atomic numbers. Since efficiency depends significantly on quenching for low energy electrons, a wide energy interval for Auger electron affects to the accuracy of the method.

In this article we apply a KLMN four-shell model [7] which permits one to consider separately LMM, LMN and LNN Auger electrons. Such a model generates 262 different pathways, which make the determination of the probability and effective energy expressions relatively difficult. The package of programs MULTIVIA [8] permits one to obtain all these expressions by combining strings.

### 2. Counting efficiency

We obtain the counting efficiency  $\varepsilon$  for a pure electron capture nuclide from the quantities  $C$  and  $C'$

$$C = \sum_{i=1}^{240} \phi_i \exp\left(\frac{-E_i}{\lambda}\right) + \sum_{i=241}^{262} \phi_i \exp\left(\frac{-E_i}{\lambda}\right) \int_0^{\nu_i} \exp\left(-\frac{T(E, \nu_i)}{\lambda}\right) dE, \quad (1)$$

$$C' = \sum_{i=1}^{240} \phi_i \exp\left(\frac{-E_i}{2\lambda}\right) + \sum_{i=241}^{262} \phi_i \exp\left(\frac{-E_i}{2\lambda}\right) \int_0^{\nu_i} \exp\left(-\frac{T(E, \nu_i)}{2\lambda}\right) dE, \quad (2)$$

where  $i$  denotes each of the 262 different ways of atomic rearrangement,  $\phi_i$  is the rearrangement probability and  $T(E, \nu_i)$  is the reduced spectrum for Compton and photoelectric electrons. We define  $\nu_i$  as follows:

$$\nu_{240} = E_{KN}, \quad \nu_{242} = \nu_{244} = \nu_{245} = E_{KM}, \quad \nu_{243} = \nu_{246} = \dots = \nu_{262} = E_{KL}. \quad (3)$$

The quantities  $C$  and  $C'$  are related to the counting efficiency by expression

$$\varepsilon = 1 - 2C' + C. \quad (4)$$

### 3. Probabilities and effective energies for the pathways

In a KLMN four-shell model, the electron capture process creates a hole in one of the three shells K, L or M. This hole leads to the emission of one X-ray or Auger electron, which generates new vacancies in higher levels. The new holes are filled again by a cascade of electrons that shifts holes to higher levels. We can achieve all atomic rearrangement pathways by only combining the two annihilation-creation

operators Auger and X-ray. The Auger operator  $A(X)$  annihilates a hole in X-shell, but creates two new holes in outer shells. The X-ray operator  $X(X)$  shifts a hole in X-shell to a higher level.

We can obtain all possible pathways in four steps. In the first step, we do not apply any of the two operators Auger or X-ray. We start with a set of holes  ${}^1B$  generated by the capture of one electron in K, L or M shells.

$${}^1B = \{K, L, M\}. \quad (5)$$

In the next steps, we apply Auger and X-ray operators to all possible holes. The second step conforms the set  ${}^2B$  of 22 events, which we list below

$${}^2B = \{A(K) = LL, LM, LN, MM, MN, NN; \quad A(L) = MM, MN, NN; \quad A(M) = NN; \\ X(K) = L_d, L_u, M_d, M_u, N_d, N_u; \quad X(L) = M_d, M_u, N_d, N_u; \quad X(N) = N_d, N_u\}. \quad (6)$$

The subscripts d and u denote detection and non-detection for X-rays, respectively. The third and fourth steps shift out holes to M- and N-shells and conform a final set  ${}^4B$  of 612 events.

The probability for each event in the final set  ${}^4B$  is a Markov chain

$$P(B_i) = P({}^1B_{i_1}) P({}^2B_{i_2}/{}^1B_{i_1}) P({}^3B_{i_3}/{}^2B_{i_2}) P({}^4B_{i_4}/{}^3B_{i_3}), \quad (7)$$

where  $B_i$  refers to all final events while  $B_{i_1}$ ,  $B_{i_2}$ ,  $B_{i_3}$  and  $B_{i_4}$  represent all events into the four sets  ${}^1B$ ,  ${}^2B$ ,  ${}^3B$  and  ${}^4B$ .

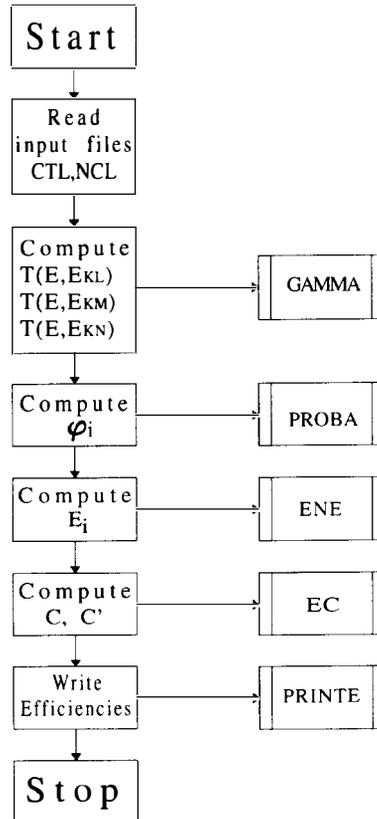


Fig. 1. Structure of the program CAPMULT.

Due to the presence of symmetries, many of the computed events have the same probability. We can reduce the total number of events to 262 pathways by assigning a weight to each expression.

Auger and X-ray operators do not operate on a hole with the same probability. Therefore each probability  $P(B_{i_j}/B_{i_{j-1}})$  must include the fluorescence yield or the Auger emission probability. Also the two possibilities for X-rays, detection or non-detection, require the computation of the escape probabilities for KL, KM, KN, LM, LN and MN photons.

Finally, we can deduce the effective energy expressions for the 262 pathways by applying a conversion of characters to all probability expressions.

#### 4. Program structure

The program CAPMULT contains a main program and 13 subprograms. Fig. 1 illustrates the essential structure of the program. The subroutine GAMMA computes the escape probabilities for KL, KM, KN, LM, LN, MN photons and the Compton distributions  $T(E, \nu_i)$  for KL, KM and KN photons [6]. The subroutine PROBA includes the 262 expressions we need for the computation of the probabilities while the subroutine ENE lists their respective effective energies. Finally, the achievement of the counting efficiency for the nuclide requires the subroutine EC, which computes the quantities  $C$  and  $C'$  (1, 2).

#### 5. Input-output data files

The input data file NCL contains the following atomic and nuclear data for the nuclide:

WK, WL, WM	$\omega_K, \omega_L$ and $\omega_M$ fluorescence yields,
PKLL, PKLM, PKLN, ...	
PLMM, PLMN, PLNN	
PMNN	Auger probabilities,
PKL, PKM, PKN	
PLM, PLN	

Table 1  
Listing of nuclide data file NCL

T1204	
''	ATOMIC DATA
'WK, ''WL, ''WM	: ' .966, '' .3434, '' .0260
'PKLL, ''PKLM, ''PKLN...	: ' .6078, '' .2640, '' .0736, '' .03, '' .0164, '' .0182
'PLMM, ''PLMN, ''PLNN	: ' .5658, '' .3721, '' .0621
'PMNN	: ' 1.
'EKLL, ''EKLM, ''EKN...	: ' 55.5, ''66.4, ''68.8, ''77.3, ''79.7, ''82.1
'ELMM, ''ELMN, ''ELNN	: ' 8., ''10.4, ''12.8
'EMNN	: ' 1.9
'PKL, ''PKM, ''PKN	: ' .7825, '' .1702, '' .0473
'PLM, ''PLN	: ' .7080, '' .2920
'PMN	: ' 1.
'EKL, ''EKM, ''EKN	: ' 69.3, ''80.2, ''82.6
'ELM, ''ELN	: ' 10.9, ''13.3
'EMN	: ' 2.4
''	NUCLEAR DATA
'PK, ''PL, ''PM	: ' .6459, '' .2740, '' .0801

Table 2  
Listing of control data file CTL

	CONTROL DATA	
'R,'"H,'"NSUC	: '	1.25,'"3.05,'"2000
'FIN,'"FFIN,'"DINC	: '	1.,'"16.0,'"1
'SCINTILLATOR	: '	5

PMN	X-ray probabilities,
EKLL, EKLM, EKLN, ...	
ELMM, ELMN, ELNN	
EMNN	Auger energies,
EKL, EKM, EKN	
ELM, ELN	
EMN	X-ray energies,
PK, PL, PM	EC probabilities.

The input data file CTL includes the internal vial radius and vial height, the number of Monte Carlo simulating X-ray photons, the free parameter interval and increment, and the scintillator parameter  $l$ , where

$l = 1$	Toluene,
$l = 2$	Instagel,
$l = 3$	Hisafe II,
$l = 4$	Dioxane-Naphthalene,
$l = 5$	Ultima-Gold.

The counting efficiencies are finally listed in the output data file EFF.

## 6. Test run

The Test Run Output at the end of this paper lists the counting efficiencies for the capture branch of  $^{204}\text{Tl}$  when 15 ml of Ultima-Gold are used. The input data files CTL and NCL are shown in Tables 1 and 2.

## References

- [1] A. Grau Malonda, Appl. Rad. Isotop. 33 (1982) 371.
- [2] J.M. Los Arcos, A. Grau Malonda and A. Fernandez, Comput. Phys. Commun. 44 (1987) 209.
- [3] E. Günther and A. Grau Carles, to be published.
- [4] A. Grau Carles, Appl. Rad. Isotop. 45 (1994) 655.
- [5] A. Grau Carles, A. Grau Malonda and L. Rodriguez Barquero, Appl. Rad. Isotop. 45 (1994) 461.
- [6] A. Grau Carles, A. Grau Malonda and P. Grau Carles. Comput. Phys. Commun. 79 (1994) 115.
- [7] G. Galiano Casas and A. Grau Malonda, CIEMAT Report 742 (Madrid, 1994).
- [8] G. Galiano Casas, to be published.

## TEST RUN OUTPUT

COUNTING EFFICIENCY FOR T1204

F. PARAM	EFFICIENCY	F. PARAM	EFFICIENCY	F. PARAM	EFFICIENCY
1.00	0.7507 **	6.00	0.2512 **	11.00	0.1558 **
1.10	0.7370 **	6.10	0.2475 **	11.10	0.1549 **
1.20	0.7226 **	6.20	0.2439 **	11.20	0.1540 **
1.30	0.7077 **	6.30	0.2405 **	11.30	0.1532 **
1.40	0.6923 **	6.40	0.2372 **	11.40	0.1524 **
1.50	0.6765 **	6.50	0.2340 **	11.50	0.1515 **
1.60	0.6605 **	6.60	0.2308 **	11.60	0.1507 **
1.70	0.6443 **	6.70	0.2278 **	11.70	0.1500 **
1.80	0.6282 **	6.80	0.2249 **	11.80	0.1492 **
1.90	0.6122 **	6.90	0.2221 **	11.90	0.1485 **
2.00	0.5964 **	7.00	0.2194 **	12.00	0.1477 **
2.10	0.5809 **	7.10	0.2167 **	12.10	0.1470 **
2.20	0.5657 **	7.20	0.2142 **	12.20	0.1463 **
2.30	0.5508 **	7.30	0.2117 **	12.30	0.1456 **
2.40	0.5363 **	7.40	0.2093 **	12.40	0.1450 **
2.50	0.5223 **	7.50	0.2070 **	12.50	0.1443 **
2.60	0.5087 **	7.60	0.2047 **	12.60	0.1437 **
2.70	0.4955 **	7.70	0.2025 **	12.70	0.1430 **
2.80	0.4827 **	7.80	0.2004 **	12.80	0.1424 **
2.90	0.4704 **	7.90	0.1983 **	12.90	0.1418 **
3.00	0.4586 **	8.00	0.1963 **	13.00	0.1412 **
3.10	0.4471 **	8.10	0.1944 **	13.10	0.1406 **
3.20	0.4361 **	8.20	0.1925 **	13.20	0.1401 **
3.30	0.4255 **	8.30	0.1906 **	13.30	0.1395 **
3.40	0.4153 **	8.40	0.1888 **	13.40	0.1390 **
3.50	0.4055 **	8.50	0.1871 **	13.50	0.1384 **
3.60	0.3960 **	8.60	0.1854 **	13.60	0.1379 **
3.70	0.3870 **	8.70	0.1838 **	13.70	0.1374 **
3.80	0.3782 **	8.80	0.1822 **	13.80	0.1369 **
3.90	0.3699 **	8.90	0.1806 **	13.90	0.1364 **
4.00	0.3618 **	9.00	0.1791 **	14.00	0.1359 **
4.10	0.3540 **	9.10	0.1776 **	14.10	0.1354 **
4.20	0.3466 **	9.20	0.1762 **	14.20	0.1349 **
4.30	0.3394 **	9.30	0.1748 **	14.30	0.1345 **
4.40	0.3325 **	9.40	0.1735 **	14.40	0.1340 **
4.50	0.3259 **	9.50	0.1721 **	14.50	0.1336 **
4.60	0.3195 **	9.60	0.1708 **	14.60	0.1332 **
4.70	0.3133 **	9.70	0.1696 **	14.70	0.1327 **
4.80	0.3074 **	9.80	0.1684 **	14.80	0.1323 **
4.90	0.3017 **	9.90	0.1672 **	14.90	0.1319 **
5.00	0.2962 **	10.00	0.1660 **	15.00	0.1315 **
5.10	0.2910 **	10.10	0.1649 **	15.10	0.1311 **
5.20	0.2859 **	10.20	0.1638 **	15.20	0.1307 **
5.30	0.2810 **	10.30	0.1627 **	15.30	0.1303 **
5.40	0.2762 **	10.40	0.1616 **	15.40	0.1299 **
5.50	0.2717 **	10.50	0.1606 **	15.50	0.1296 **
5.60	0.2673 **	10.60	0.1596 **	15.60	0.1292 **
5.70	0.2630 **	10.70	0.1586 **	15.70	0.1288 **
5.80	0.2590 **	10.80	0.1577 **	15.80	0.1285 **
5.90	0.2550 **	10.90	0.1567 **	15.90	0.1281 **