EMI2, the counting efficiency for electron capture by a $KL_1L_2L_3M$ model

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Abstract

The program EMI2 computes the liquid-scintillation counting efficiency for pure electron capture, single isomeric and electron capture-gamma nuclides. In this second version, the atomic rearrangement process subsequent to electron capture is described in terms of a more sophisticated $KL_1L_2L_3M$ model of 264 different pathways. The addition of Coster–Kronig transitions improves the accuracy of the method, especially for nuclides of low atomic numbers. A Monte Carlo method is applied to simulate the photoelectric and Compton interactions of X- and γ -photons with the scintillator inside the vial geometry.

Keywords: Radioactivity; Liquid scintillation counting; Electron capture; Coster–Kronig emission

NEW VERSION SUMMARY

Title of program: EMI2

Catalogue identifier: ADKR

Program Summary URL:
http://www.cpc.cs.qub.ac.uk/cpc/summaries/ADKR

Program obtainable from: CPC Program Library, Queen's University of Belfast, N. Ireland

Computers: any IBM compatible with 80386 or higher Intel processor

Operating systems under which the program has been tested: MS-DOS and higher systems

Previous version: ACPU, Comput. Phys. Commun. 79 (1994) 115

Programming language used: FORTRAN 77

Memory required to execute with typical data: 30 kwords

No. of bits in a word: 32

No. of bytes in distributed program, including test data, etc.:
119 955

Distribution format: ASCII

Keywords: Radioactivity, liquid scintillation counting, electron capture, Coster–Kronig emission

Nature of physical problem
The standardization of electron capture (EC) nuclides in liquid scintillation detectors is never 100% efficient, depending on several factors such as ionization or chemical quench. A complete description

of the EC process is not possible to achieve due to the large number of involved atomic rearrangement pathways. In the first version of the program EMI, an oversimplified *KLM* model of only 22 pathways was applied. However, a better accuracy in the standardizations, especially for nuclides of low atomic numbers, requires one to include Coster–Kronig transitions, and therefore L_i-subshells.

Method of solution
The atomic rearrangement process for the $KL_1L_2L_3M$ model is described by a set of 264 different equations. This number suggests the convenience of a code for computing all equations.

Restrictions on the complexity of the problem
We assume that the five M subshells are averaged. The atomic rearrangement for M and higher shells is not considered.

Typical running time
The test run requires about 15 s on a Pentium II PC.

LONG WRITE-UP

1. Introduction

Early programs [1,2] to compute the liquid scintillation counting efficiency for electron-capture nuclides involved a crude model of only three shells. Additionally, the interaction of X-rays with the liquid scintillator was assumed to be only photoelectric. Further experimental work proved that the photoelectric approximation was clearly insufficient [3,4], and a first version of program EMI [5] was developed to include the Compton contribution of X-rays.

Last international comparisons for electron capture nuclides [6,7] revealed the interest of the generalization of the primitive *KLM* three-shell model applied to calibrate nuclides by the CIEMAT/NIST method [8–10]. A new *KLMN* four-shell model [11,12] was proposed to consider *LMM*, *LMN* and *LNN* Auger electrons separately. This model increased the number of involved probability and energy equations from 21 to 262. For such a reason, the development of a computer routine to derive the complete set of equations is of great interest. Calculations were based on strings which could be modified by the application of creation and annihilation operators [13]. The *KLMN* model showed a better agreement between computed and experimental efficiencies for nuclides of high atomic numbers [14].

A substantial improvement of the method for low atomic number nuclides, e.g., ^{55}Fe, consists of adding the three atomic subshells L_1, L_2 and L_3. Once again the large number of involved equations (this time 264) makes suitable the application of strings and annihilation-creation operators. The $KL_1L_2L_3M$ model requires one to add the operator Coster–Kronig to the two above-mentioned Auger and X-ray operators.

The program EMI2 computes the liquid scintillation counting efficiency for nuclides of different decay schemes: electron capture, single isomeric transitions and electron capture followed by coincident or non-coincident gamma transitions. Since the interaction of γ- and X-rays depends on the composition of the scintillator, the atomic cross sections must be computed separately for the different commercially available scintillators, e.g., toluene, Insta-Gel, Hisafe II or Ultima-Gold. Also ionization quench is an important parameter that can modify the counting efficiency. The program EMI2 applies a six-coefficient equation to fit the semiempirical equation of Birks for the ionization quench factor $Q(E)$. The six coefficients are interpolated linearly in a table of 20 increasing values of kB [16,17].

2. The counting efficiency

The computation of the counting efficiency for pure electron capture nuclides, coincident and non-coincident electron capture-gamma transitions and single isomeric transitions can be considerably simplified if we make use

of the parameters $C, C', \Sigma, \Sigma', \Gamma$ and Γ' [5]. This notation permits one to write the counting efficiency for pure electron capture nuclides as follows:

$$\varepsilon_{\mathrm{EC}} = 1 - 2C' + C, \tag{1}$$

where

$$C = \sum_{i=1}^{255} \phi_i \exp\left(-\frac{E_i}{\lambda}\right) + \sum_{i=256}^{265} \phi_i \exp\left(-\frac{E_i}{\lambda}\right) \int_0^{\nu_i} \exp\left(\frac{T(E,\nu_i)}{\lambda}\right) dE, \tag{2}$$

$$C' = \sum_{i=1}^{255} \phi_i \exp\left(-\frac{E_i}{2\lambda}\right) + \sum_{i=256}^{265} \phi_i \exp\left(-\frac{E_i}{2\lambda}\right) \int_0^{\nu_i} \exp\left(\frac{T(E,\nu_i)}{2\lambda}\right) dE. \tag{3}$$

The subindex i denotes each of the 264 different pathways of atomic rearrangement, ϕ_i is the probability of each pathway and $T(E,\nu_i)$ is the reduced energy spectrum for Compton and photoelectric electrons. The parameter ν_i can take the values

$$\nu_{256} = E_{KM}, \qquad \nu_{257} = \nu_{259} = \nu_{260} = \nu_{264} = \nu_{265} = E_{KL_2}, \qquad \nu_{258} = \nu_{261} = \nu_{262} = E_{KL_3}. \tag{4}$$

The counting efficiency for single isomeric transitions ε_{IT} is computed as follows:

$$\varepsilon_{IT} = p_{IC}\varepsilon_{IC} + p_\gamma \varepsilon_\gamma, \tag{5}$$

where p_{IC} and p_γ are the internal conversion and the gamma-ray emission probabilities, respectively. The internal conversion efficiency ε_{IC} is given by

$$\varepsilon_{IC} = 1 - 2\Sigma' + \Sigma, \tag{6}$$

where

$$\Sigma = \sum_{i=1}^{255} w_i \exp\left(-\frac{E_i'}{\lambda}\right) + \sum_{i=236}^{265} w_i \exp\left(-\frac{E_i'}{\lambda}\right) \int_0^{\nu_i} \exp\left(-\frac{T(E',\nu_i)}{\lambda}\right) dE, \tag{7}$$

$$\Sigma' = \sum_{i=1}^{255} w_i \exp\left(-\frac{E_i'}{2\lambda}\right) + \sum_{i=236}^{265} w_i \exp\left(-\frac{E_i'}{2\lambda}\right) \int_0^{\nu_i} \exp\left(-\frac{T(E',\nu_i)}{2\lambda}\right) dE. \tag{8}$$

Although Eqs. (2), (3) and (7), (8) may look similar in structure, they describe different physical processes. The probability ϕ_i describes the capture process of one electron by the nucleus, while the probability w_i considers the ejection of one electron away from an atomic level. The clear analogy between the two equations results from the atomic rearrangement that follows from both processes. The prime on energy E_i' expresses that the kinetic energy of the converted electron is taken into account. The efficiency ε_γ for gamma-ray emission is given by

$$\varepsilon_\gamma = p_\gamma(1 - I_\gamma)(1 - 2\Gamma + \Gamma'), \tag{9}$$

where p_γ is the gamma-ray emission probability and I_γ is the escape probability when gamma rays do not suffer interaction with the detection system. The parameters Γ and Γ' are given by

$$\Gamma = \int_0^{E_\gamma} \exp\left(-\frac{T(E,E_\gamma)}{\lambda}\right) dE, \tag{10}$$

$$\Gamma = \int_0^{E_\gamma} \exp\left(-\frac{T(E,E_\gamma)}{2\lambda}\right) dE, \tag{11}$$

respectively. Both E_γ and E are reduced energies.

The counting efficiency for electron capture nuclides coincident with gamma transitions is computed from equation

$$\varepsilon = p_{IC}(1 - 2C\Sigma + C'\Sigma') + p_\gamma(1 - I_\gamma)(1 - 2C\Gamma + C'\Gamma') + p_\gamma I_\gamma(1 - 2C + C'). \tag{12}$$

3. Probability and effective energy

The model $KL_1L_2L_3M$ considers atomic rearrangement processes in which electron capture or internal conversion creates vacancies in K- or L_i-shells. When we average the three L_i-subshells to one L-shell, these vacancies only lead to the emission of X-ray or Auger electrons. However, when the holes are produced in L_1 and L_2-subshells, they are followed by the so-called Coster–Kronig transitions. The process creates a cascade of electrons that shifts holes to higher levels. We may define the three annihilation-creation operators, Auger $A(X)$, X-ray $X(X)$ and Coster–Kronig $C(X)$. The Auger operator $A(X)$ annihilates a hole in the X-shell, but creates two new holes in the outer shells. The Coster–Kronig operator $C(X)$ annihilates a hole in L_1 or L_2 subshell, but creates two new holes in L_i- and M-shells. The X-ray operator $X(X)$ shifts a hole from the X-shell to a higher level.

We may compute all possible pathways from strings in five steps. The first step considers a set of holes ${}^1B$ generated by the capture of one electron from the K-shell or L_1, L_2 and L_3-subshells,

$${}^1B = \{K, L_1, L_2, L_3\}. \tag{13}$$

The following steps apply Auger, X-ray and Coster–Kronig operators to the holes produced by the capture process. The second step conforms a set ${}^2B$ of 30 events, which are listed below:

$$\begin{aligned}
{}^2B = \big\{ & A(K) = L_1L_1, L_1L_2, L_1L_3, L_2L_2, L_2L_3, L_3L_3, L_1M, L_2M, L_3M, MM; \\
& A(L_1) = MM; \qquad A(L_2) = MM; \qquad A(L_3) = MM; \\
& X(L_1) = M_d, M_u; \qquad X(L_2) = M_d, M_u; \qquad X(L_3) = M_d, M_u; \\
& C(L_1) = L_2M, L_3M; \qquad C(L_2) = L_3M; \\
& X(K) = L_{1d}, L_{1u}, L_{2d}, L_{2u}, L_{3d}, L_{3u}, M_d, M_u \big\}.
\end{aligned} \tag{14}$$

The subscripts d and u denote detection and non-detection of X-rays, respectively. The third, fourth and fifth steps shift holes to L_2-, L_3- and M-; L_3- and M-; and M-shell, respectively, and finally conform a set ${}^5B$ of 378 events.

The probability of each event into the final set ${}^5B$ is the following Markov chain:

$$P(B_i) = P\left({}^1B_{i_1}\right) P\left({}^2B_{i_2}/{}^2B_{i_1}\right) P\left({}^3B_{i_3}/{}^2B_{i_2}\right) P\left({}^4B_{i_4}/{}^2B_{i_3}\right) P\left({}^5B_{i_5}/{}^4B_{i_4}\right), \tag{15}$$

where B_{i_1}, B_{i_2}, B_{i_3}, B_{i_4} and B_{i_5} are events into the five sets ${}^1B$, ${}^2B$, ${}^3B$, ${}^4B$ and ${}^5B$.

The presence of symmetries makes many of the computed events to have the same probability. The total number of events can be reduced to 264 pathways by assigning the adequate weights to all expressions.

Auger, X-ray and Coster–Kronig operators do not operate on a hole with the same probability, and each probability $P(B_{i_j}/B_{i_{j-1}})$ must include fluorescence, Auger or Coster–Kronig yields. Also the two possible events of detection or non-detection for X-rays require the computation of the escape probabilities for $KL_1, KL_2, KL_3, KM, L_1M, L_2M$ and L_3M photons.

The reduced energies are computed by multiplying energy E by the ionization quench factor

$$Q(E) = \frac{A_1 + A_2 \ln E + A_3(\ln E)^2 + A_4(\ln E)^3}{1 + A_5 \ln E + A_6(\ln E)^2 + A_4(\ln E)^3}, \tag{16}$$

where A_i $(i = 1, 6)$ are tabulated coefficients [17] which depend on the kB value.

4. Program structure

The program EMI2 contains a main program and 13 subprograms. Fig. 1 illustrates the basic structure of the program. The subroutine GAMMA computes all escape probabilities for KL_1, KL_2, KL_3, KM, L_1M, L_2M, L_3M photons and the Compton distributions $T(E, \nu_i)$ for KL_1, KL_2, KL_3 and KM photons. The subroutines PROBA and ENE1 contain the list of 264 different pathway equations for the probabilities and energies. The counting efficiency parameters $C, C', \Sigma, \Sigma', \Gamma$ and Γ' are computed in the subroutines EC, IC or GAM, depending on the decay scheme of the nuclide. The subroutines PUNTO, DIREC generate a random point and give a random direction for the photon inside the vial geometry. The function DADO generates a random number into the range between 0 and 1 [18]. The subroutine SECC computes the photoelectric and Compton cross sections for a given scintillator. Finally, the mean path length for the photon is derived in the subroutine ESPEC.

5. Input–output data files

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

PK, PL1, PL2, ...	EC probabilities
PIK, PIL1, PIL2, ...	IC probabilities
WK, WL1, WL2, WL3	Fluorescence yield
F12, F13, F23	Coster–Kronig yield
PKL1L1, PKL1L2...	K-Auger probabilities
PL1L2M, PL1L3M...	Coster–Kronig probabilities
PL1MM, PL2MM, PL3MM	L-Auger probabilities
EKL1L2, EKL1L2...	K Auger energies
EL1L2M, EL1L3M...	Coster–Kronig energies
EL1MM, EL2MM, EL2MM	L Auger energies
PKL2, PKL3, PKM	K X-ray probabilities
PL1M, PL2M, PL3M	L X-ray probabilities
EKL2, EKL3, EKM	K X-ray energies
EL1M, PL2M, PL3M	L X-ray energies
EGAM	Gamma-ray energy
PGAM	Gamma-ray probability

The file CTL includes the internal radius of the vial and its height, the number of Monte-Carlo simulating X- and γ-ray photons, the free parameter interval and increment, the kB value for ionization quench correction, and the scintillator parameter l, which takes the values

$l = 1$ Toluene

$l = 2$ InstaGel

$l = 3$ Hisafe II

$l = 4$ Dioxane-Naphtalene

$l = 5$ Ultima-Gold

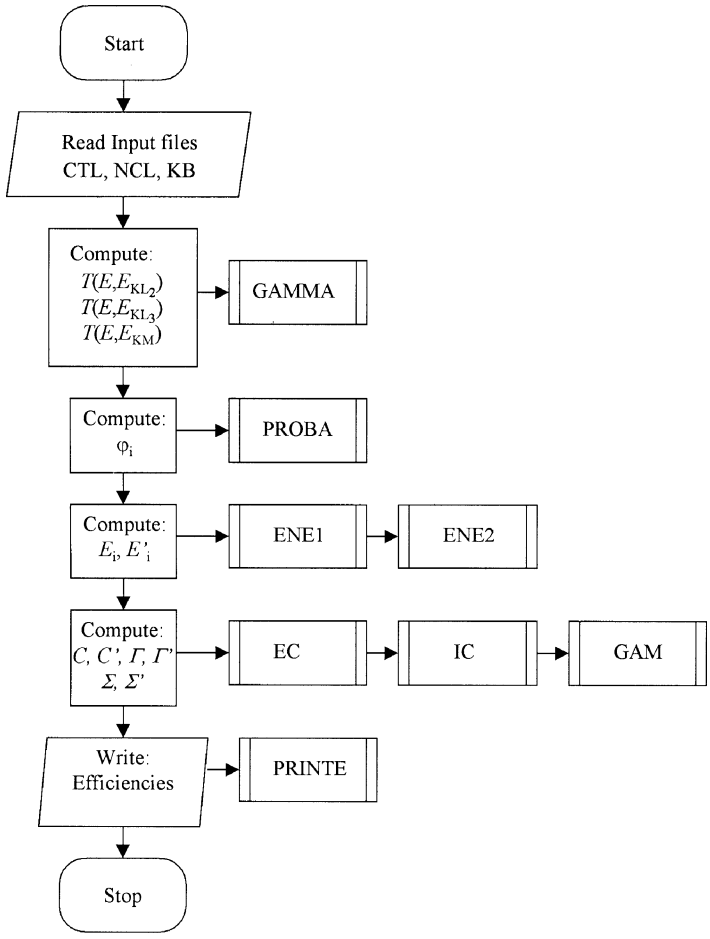

Fig. 1. Structure of the program EMI2.

```
''                      CONTROL DATA
'DECAY SCHEME                  :'          4
'R,H,NSUC                      :'          1.25,3.05,2000
'FIN,FFIN,DINC                 :'          1.,8.5,.05
'SCINTILLATOR                  :'          5
'KB                            :'          0.0075
```

Fig. 2. Listing of control data file CTL.

The counting efficiencies are printed in the output data file EFF.TAB.

6. Test run

As a test run we have computed the counting efficiency for ^{125}I, when the free parameter is in the interval between 1 and 8.5. Figs. 2, 3 and 4 show the input data files CTL, NCL and KB, respectively.

```
                          'I125'
''                      ATOMIC DATA
'WK,WL1,WL2,WL3            :'          .877,.041,.074,.0074
'F12,F13,F23               :'          .18,.28,.155
'PKL1L1,PKL1L2 ...         :'          .0777,.0926,.1154,.0119,.2520,.1282
                                       .0849,.0781,.1326,.0266
'PL1L2M,PL1L3M,PL2L3M      :'          1.,1.,1.
'PL1MM,PL2MM,PL3MM         :'          1.,1.,1.
'EKL1L1,EKL1L2 ...         :'      21.935,22.262,22.533,22.589,22.860,23.131
                                       26.166,26.510,26.760,30.276
'EL1LL2M,EL1L3M,EL2L3M     :'          .2208,.5033,.1963
'EL1MM,EL2MM,EL3MM         :'          3.592,3.525,3.138
'PKL2,PKL3,PKM             :'          .2898,.5355,.1747
'PL1M,PL2M,PL3M            :'          1.,1.,1.
'EKL2,EKL3,EKM             :'          27.201,27.472,31.102
'EL1M,EL2M,EL3M            :'          4.240,4.077,3.829
'EK,EL1,EL2,EL3,EM         :'          31.814,4.939,4.612,4.341,1.01

''                      NUCLEAR DATA
'PK,PL1,PL2,PL3,PM         :'          .806,.157,.004,.0,.033
'PGAM,EGAM                 :'          .0658,35.49
'PIK,PIL1,PIL2, ...        :'          .8608,.1032,.0082,.0020,.0255
```

Fig. 3. Listing of nuclide data file NCL.

'KB	A1	A2	A3	A4	A5	A6'
0.001	.848599	.385794	.155281	-5.91812E-3	.389966	0.151436
0.002	.739162	.338152	.141140	-4.12402E-3	.345868	0.134519
0.003	.655854	.304306	.131988	-4.75264E-3	.313821	0.123448
0.004	.589173	.264733	.121728	-2.54649E-3	.273885	0.111985
0.005	.535514	.239700	.114018	-1.95557E-3	.249569	0.103179
0.006	.490419	.216002	.107251	-1.46804E-3	.224681	0.954474E-1
0.007	.453549	.202317	.101371	-0.79200E-3	.214455	0.888047E-1
0.008	.421495	.187310	.962573E-1	-0.52346E-3	.199778	0.831139E-1
0.009	.394398	.173722	.912972E-1	-0.57124E-3	.185241	0.778465E-1
0.010	.368515	.163988	.881641E-1	-0.45562E-3	.175826	0.743192E-1
0.011	.347937	.151931	.842714E-1	-0.55260E-4	.164438	0.700270E-1
0.012	.329035	.144041	.805924E-1	-0.45360E-4	.156239	0.661165E-1
0.013	.312306	.136647	.774480E-1	-0.43060E-4	.149609	0.628423E-1
0.014	.296689	.128723	.747768E-1	0.15992E-3	.140914	0.597828E-1
0.015	.282808	.122593	.723592E-1	0.30371E-3	.135895	0.572500E-1
0.016	.270204	.116638	.698574E-1	0.34938E-3	.129336	0.545388E-1
0.017	.258631	.111339	.676496E-1	0.41188E-3	.124146	0.521832E-1
0.018	.248080	.106463	.655764E-1	0.46844E-3	.119382	0.499835E-1
0.019	.238321	.101925	.636383E-1	0.51606E-3	.114937	0.479306E-1
0.020	.229338	.097848	.618485E-1	0.56027E-3	.110961	0.460418E-1

Fig. 4. Ionization quench coefficients for different KB values.

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TEST RUN OUTPUT

```
COUNTING EFFICIENCY FOR I125
-----------------------------------------------------------------
F.PARAM  EFFICIENCY   F.PARAM  EFFICIENCY   F.PARAM  EFFICIENCY
-------  ----------   -------  ----------   -------  ----------
  1.00     .9039  **    3.50     .5986  **    6.00     .4574  **
  1.05     .8951  **    3.55     .5950  **    6.05     .4551  **
  1.10     .8863  **    3.60     .5914  **    6.10     .4529  **
  1.15     .8775  **    3.65     .5879  **    6.15     .4506  **
  1.20     .8687  **    3.70     .5844  **    6.20     .4484  **
  1.25     .8599  **    3.75     .5810  **    6.25     .4462  **
  1.30     .8512  **    3.80     .5776  **    6.30     .4440  **
  1.35     .8426  **    3.85     .5743  **    6.35     .4419  **
  1.40     .8341  **    3.90     .5710  **    6.40     .4397  **
  1.45     .8257  **    3.95     .5677  **    6.45     .4375  **
  1.50     .8174  **    4.00     .5645  **    6.50     .4354  **
  1.55     .8094  **    4.05     .5613  **    6.55     .4333  **
  1.60     .8014  **    4.10     .5582  **    6.60     .4312  **
  1.65     .7936  **    4.15     .5550  **    6.65     .4291  **
  1.70     .7860  **    4.20     .5520  **    6.70     .4270  **
  1.75     .7785  **    4.25     .5489  **    6.75     .4250  **
  1.80     .7713  **    4.30     .5459  **    6.80     .4229  **
  1.85     .7641  **    4.35     .5429  **    6.85     .4209  **
  1.90     .7572  **    4.40     .5399  **    6.90     .4189  **
  1.95     .7504  **    4.45     .5370  **    6.95     .4169  **
  2.00     .7437  **    4.50     .5341  **    7.00     .4149  **
  2.05     .7372  **    4.55     .5312  **    7.05     .4129  **
  2.10     .7309  **    4.60     .5284  **    7.10     .4109  **
  2.15     .7247  **    4.65     .5255  **    7.15     .4090  **
  2.20     .7186  **    4.70     .5227  **    7.20     .4070  **
  2.25     .7127  **    4.75     .5200  **    7.25     .4051  **
  2.30     .7069  **    4.80     .5172  **    7.30     .4032  **
  2.35     .7013  **    4.85     .5145  **    7.35     .4013  **
  2.40     .6958  **    4.90     .5118  **    7.40     .3994  **
  2.45     .6904  **    4.95     .5091  **    7.45     .3975  **
  2.50     .6851  **    5.00     .5065  **    7.50     .3956  **
  2.55     .6800  **    5.05     .5038  **    7.55     .3938  **
  2.60     .6749  **    5.10     .5012  **    7.60     .3919  **
  2.65     .6700  **    5.15     .4986  **    7.65     .3901  **
  2.70     .6651  **    5.20     .4960  **    7.70     .3883  **
  2.75     .6604  **    5.25     .4935  **    7.75     .3865  **
  2.80     .6557  **    5.30     .4910  **    7.80     .3847  **
  2.85     .6512  **    5.35     .4884  **    7.85     .3829  **
  2.90     .6467  **    5.40     .4859  **    7.90     .3811  **
  2.95     .6423  **    5.45     .4835  **    7.95     .3794  **
  3.00     .6380  **    5.50     .4810  **    8.00     .3776  **
  3.05     .6337  **    5.55     .4786  **    8.05     .3759  **
  3.10     .6296  **    5.60     .4762  **    8.10     .3742  **
  3.15     .6255  **    5.65     .4737  **    8.15     .3725  **
  3.20     .6215  **    5.70     .4714  **    8.20     .3708  **
  3.25     .6175  **    5.75     .4690  **    8.25     .3691  **
  3.30     .6136  **    5.80     .4666  **    8.30     .3674  **
  3.35     .6098  **    5.85     .4643  **    8.35     .3657  **
  3.40     .6060  **    5.90     .4620  **    8.40     .3640  **
  3.45     .6023  **    5.95     .4597  **    8.45     .3624  **
-----------------------------------------------------------------
```