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Internal Conversion Coefficients - How good are they now?

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Abstract. Internal conversion coefficients involving atomic electrons (ICC) and electron–positron pairs (IPC) are often required to determine transition multiplicities and total transition rates. A new internal conversion coefficient data base, BrIcc has been developed which integrates a number of tabulations on ICC and IPC, as well as $\Omega(E0)$ electronic factors. To decide which theoretical internal conversion coefficient table to use the accurately determined experimental α_K , α_L , α_{Total} and α_K/α_L values were compared with the new Dirac–Fock calculations using extreme assumptions on the effect of the atomic vacancy. While the overall difference between experiment and theory is less than 1 %, our analysis shows preference towards the so called “Frozen Orbital” approximation, which takes into account the effect of the atomic vacancy.

1 Introduction

Internal conversion coefficients (ICC) convey important information about the atomic nucleus. Through comparison of experimental ICCs with corresponding theoretical values, multiplicities and mixing ratios of nuclear transitions are determined. As well as nuclear structure research, knowledge of accurate coefficients is needed in, for example, the determination of total transition rates (required for the normalization of decay schemes), Mössbauer spectroscopy (CEMS) [1], or nuclear reaction calculations [2].

There is a long history of generation and improvements of theoretical internal conversion coefficient tables. The most recent calculations, based on the relativistic self-consistent Dirac-Fock (DF) method [3,4] represent a major advance in the improvement of the accuracy of the theoretical coefficients, which now challenges experiments at the percent level. To make the new theoretical values accessible for a very broad user community a new internal conversion coefficient data base called BrIcc has been developed [5], which is now adopted by the International Nuclear Structure and Decay Data Network [6] for all new data evaluations published in Nuclear Data Sheets.

To assess the question of “How good are the theoretical internal conversion coefficients” accurately measured values were used from time to time to carry out benchmark studies; see for example the work of Raman *et al.* [4]. This paper is focused on the critical evaluation of existing internal conversion coefficient data using rigorous statistical methods to compare adopted values with the DF values.

2 High precision experimental internal conversion coefficients

In the simplest way the ICC is defined as $\alpha = I_{CE}/I_\gamma$, where I_{CE} and I_γ are the electron and γ -ray intensities, respectively.

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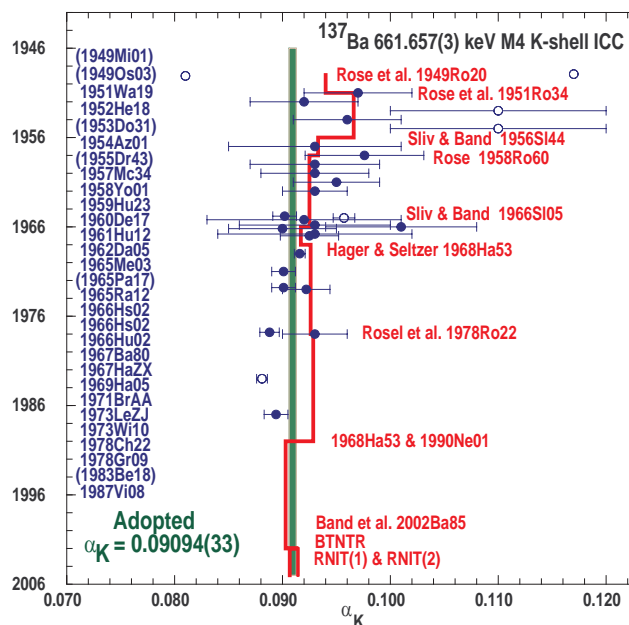


Fig. 1. Adopted value (shaded area) of the experimental α_K for the 662 keV M4 transition in ^{137}Ba . Data points: filled included in the present evaluation; open excluded. Lines (red in on-line) show the evolution of the corresponding α_{theo} values. References (experiments on the left; theory on the right) are given as NSR [7] keynumbers.

I_{CE} represents the number of electrons ejected from an atomic shell (K, L1, L2, ...), a major shell (L, M, etc.), or the sum of all shells (total), or can be the intensity of the electron–positron pairs. The internal conversion coefficient depends on the atomic number of the emitting nucleus (Z), the transition energy (E_γ), the transition multipolarity (πL) and, in the case of internal conversion, the atomic shell involved. Often the ratio of different ICCs, for example α_K/α_L or α_{L1}/α_{L2} , was used to determine the multipolarity and/or the mixing ratio.

Internal conversion coefficients also can be deduced from the intensity of X-rays or Auger electrons, which are emitted as the atomic vacancies are filled.

The measurement of internal conversion coefficients, including the methods, instrumentation and data analysis, has been developed considerably over the last 60 years, as is illustrated in Fig. 1, which shows the evolution of the experimental and theoretical K-shell internal conversion coefficient for the 661.657 keV M4 transition in ^{137}Ba . This is one of the most studied ICC; there are 29 experimental values known. In assembling the ICC data base individual data points with less than 15% experimental uncertainties have been considered; however, in the further analysis comparing experiment with theory the adopted ICC must be known to better than 5% accuracy.

We only considered α_K , α_L and α_T internal conversion coefficients and α_K/α_L ratios with $L \geq 2$ (E2, E3, M3, E4, M4 and E5) multipolarity. Electric dipole transitions are usually hindered strongly. Transitions of E1, M1 and M2 multiplicities are often mixed and the mixing ratios not always known to the accuracy needed to define the ICC values. For M1 and M2 transitions the nuclear penetration effect also could have an impact on the measured internal conversion coefficients. The determination of the multiplicities has been examined and those transitions whose multipolarity assignment is solely based on the particular ICC value were not considered. Similarly, if an ICC was used for normalization it was excluded. The transition energy and uncertainty, the multipolarity and the mixing ratio with its uncertainty (mixed transitions only) were taken from the adopted data set of the ENSDF [8].

There are a number of techniques developed to measure internal conversion coefficients [9], which usually require the knowledge of the intensity of two radiations, including conversion electrons, γ -rays, X-rays, β -rays, *etc.* In our review no preference was given to any of the methods, but in order to deduce the correct ICC significant effort went to examine every piece of information and to check if the best data were used for calibration, normalization, *etc.* For example the so called XPG method is based on the fact that each electron vacancy created in the K shell produces K X-rays with a probability equal to the K-shell fluorescent yield ω_K . The value of ω_K depends only on the atomic number, and is known with an accuracy of $\sim 0.5\%$ [10]; however, its value has changed quite significantly over the last decades. For many of the E2 transitions the total internal conversion coefficients, α_T of have been deduced by combining the reduced E2 electromagnetic transition probability, $B(E2)$, obtained from Coulomb excitation with the half-life of the level, $T_{1/2}$, from lifetime measurements. The basic formula of the so-called CEL method is defined by Raman *et al.* [4] as:

$$T_{1/2}(\text{ns}) \times (1 + \alpha_T) = \frac{2.829 \times 10^{11} \times E_\gamma^{-5}(\text{keV})}{B(E2) \uparrow (e^2 b^2)}. \quad (1)$$

Special consideration was given in deriving the uncertainties of α_T . Assuming a Gaussian distribution for both $T_{1/2}$ and $B(E2)$, $\Delta\alpha_T$ may not be Gaussian due to the inverse relationship with $T_{1/2}$ and $B(E2)$. We have adopted a procedure to estimate the uncertainty correctly taking into account this non-linear relation.

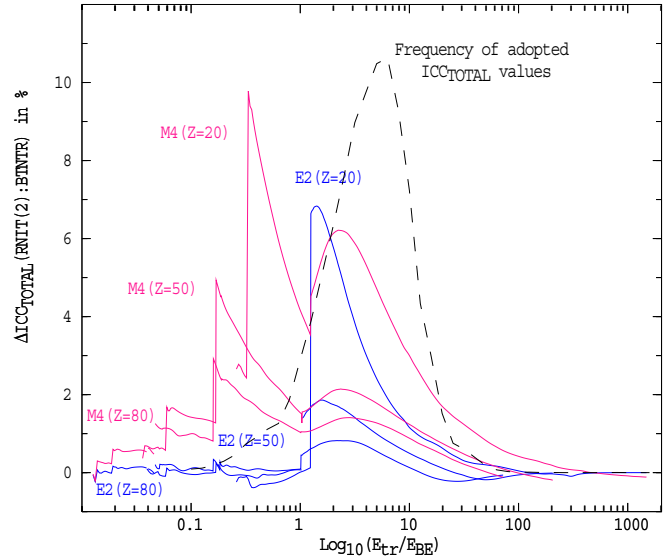


Fig. 2. Differences (in %) of the α_T values calculated using the "No Hole" (BTNTR) and "Frozen Orbital" (RNIT(2)) approximations. The horizontal axis is the transition energy (E_T) divided by the K-shell binding energy (E_{BE}). Dashed curve shows (in %) the frequency spectrum of the known α_T values in the adopted ENSDF data sets [8].

In summary, in about 22% of the cases, the ICCs (or uncertainties) have been adjusted from values originally reported. When more than one experimental value was known, adopted values were deduced using procedures described below.

There is an overlap of our data base and the two recent compilations of experimental values by Raman *et al.* [4] in 2002 and [13] in 2006. The 100 α_K and α_T values of [4] have been selected on similar principles to ours; however, a large number of corrections have been made in the verification procedures presented here. More recently [13] an extensive set of 1510 internal conversion coefficient ratios, measured with an accuracy of $\leq 10\%$ has been compiled, but no attempt was made to deduce adopted values.

3 Comparing experimental and theoretical ICCs

In comparing the experimental ICC values with a theoretical ones we have adopted the definition of [4] for the difference of $\Delta_{ICC}(\text{Exp} : \text{Theor})$, given in percentage as:

$$\Delta_{ICC}(\text{Exp} : \text{Theor}) = \frac{\alpha_i(\text{Exp}) - \alpha_i(\text{Theor})}{\alpha_i(\text{Theor})} \times 100, \quad (2)$$

where i refers to a particular ICC value of Total, K-shell, L-shell or K/L ratio, *etc.* Theoretical internal conversion coefficients, $\alpha_i(\text{Theor})$ were calculated using the original RAINE program [3] in combination with BrIcc [6]. This approach eliminates a small, $\leq 0.3\%$, systematic uncertainty of the interpolation procedure. The uncertainty of the $\alpha_i(\text{Theor})$ values has been determined from the uncertainty in transition energy and, in the case of mixed multipolarity transitions, from the uncertainty of the the mixing ratio [6].

One remaining central question concerning the theoretical ICC calculations is the treatment of atomic vacancies created

during the conversion process. The new Dirac-Fock model of Band et al. [3] offers two extreme assumptions to take into account the effect of the atomic vacancy:

- The hole is filled instantaneously, therefore the conversion process is not affected; BTNTR or "No Hole" approximation.
- The hole remains unfilled throughout the time that the conversion electron is present in the atom; RNIT(1) or "Self Consistent" approximation. An alternative approximation, RNIT(2) or "Frozen Orbital", uses a somewhat different potential in calculating the continuum wave function.

In the lack of sufficient theoretical arguments the decision to select one of the theoretical approximations must be based on careful comparison of the measured and the calculated ICCs. Fig. 2 compares the values of the total ICCs calculated with the "No Hole" and with the "Frozen Orbital" approximations and for E2 and M4 transitions in Z=20, 50 and 80. The frequency of all known α_{Total} values in the ENSDF is also shown in the figure, which can be used as a guide to illustrate the impact of the selection of the ICC data table for future evaluations.

4 Statistical treatment of the data

Three statistical techniques [11] were employed to analyze the data aiming to identify discrepant data points and deduce weighted mean values and assign uncertainties. These methods were the

- Limitation of Relative Statistical Weights Method (LWM),
- Normalized Residuals Method (NRM), and
- Rajeval Technique (RT).

If a set of data is consistent, these three methods will yield the same mean value and uncertainty and, in the case of LWM and NRM, the same reduced χ^2 . If the data set is discrepant, the results of these techniques will differ since they use different methods in treating discrepant data. These differences were used to identify deviant data points. Indications of a deviant data point could include that it was marked as "outlier" by LWM or RT; or its uncertainty was adjusted (increased) by RT or NRM. The procedure was repeated until no more deviant data point could be identified. Somewhat different procedures were used in the analysis of the experimental data and in the comparison of experiment to calculations. In the first case the adopted experimental value was taken as the arithmetic mean of the NRM and RT and the uncertainty was the larger of the NRM and RT uncertainties (providing that more than two measurements were included in the calculation). In the comparison of experiments to theory the mean value, uncertainty and reduced χ^2 of the $\Delta_{ICC}(Exp:Theor)$ differences were taken from LWM.

5 Summary

A total of 213 experimental ICC has been adopted, which are known to better than 5 % accuracy. The data set covers the Z=20 (^{44}Ca) to the Z=94 (^{240}Pu) atomic numbers and the 24 keV (^{58}Co) to 1238 keV (^{56}Fe) energy range. The total data

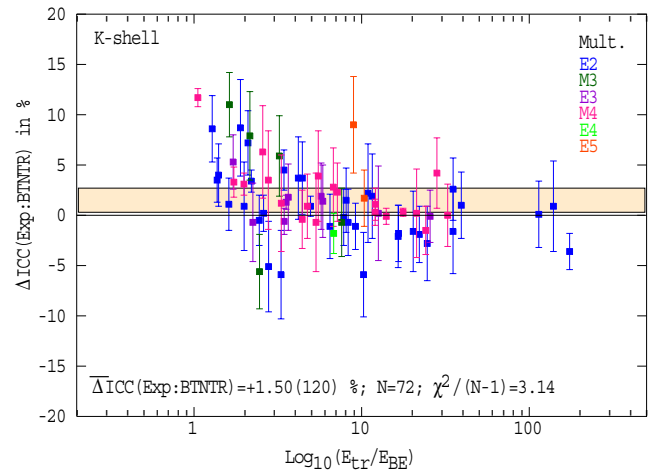


Fig. 3. Adopted value (shaded area) of the average difference between experimental α_K values and calculated ones using the so called "No Hole", BTNTR approximation.

set has been analyzed in various subgroups and compared with three different theoretical approximations, as listed in Table 1. If a $\Delta_{ICC}(Exp:Theor)$ difference for a particular experimental ICC was found to be discrepant for all three theoretical calculations, the data point was excluded from the analysis. This procedure has reduced the total number of experimental ICC values to 186. Figures 3 and 4 show in detail 72 accurately determined $\alpha_K(Exp)$ internal conversion coefficients compared with the "No Hole" and the "Frozen Orbital" approximations. The horizontal axis is the E_{tr}/E_{BE} ratio, which transforms

Table 1. Average differences (in %) between accurately measured and theoretical internal conversion coefficients.

Multi-polarity	Shell	N	$\Delta_{ICC}(Exp:Theor)$		
			BTNTR	RNIT(1)	RNIT(2)
All	All	186	+0.70(40) ^{a)}	-0.61(14)	-0.93(14)
All	T	54	+0.32(25)	-0.55(24)	-0.71(24)
All	K	72	+1.50(120) ^{a)}	-0.18(21)	-0.72(21)
All	L	9	+0.33(76)	+0.06(76)	-0.06(76)
All	K/L	46	+0.00(31)	-1.64(31)	-1.94(30)
All	T&K	126	+0.99(69) ^{a)}	-0.34(16)	-0.71(16)
E2	All	103	+0.21(23)	-0.77(23)	-0.93(23)
E2	T	36	-0.07(41)	-0.70(41)	-0.81(41)
E2	K	35	+1.06(43)	-0.04(43)	-0.25(43)
E2	K/L	23	-0.08(42)	-1.68(43)	-2.12(43)
E3	All	18	+0.83(39)	-0.69(37)	-1.07(37)
M4	All	50	+0.98(68) ^{a)}	-0.51(20)	-0.93(20)
M4	T	12	+0.40(36)	-0.65(36)	-0.82(36)
M4	K	20	+1.81(151) ^{a)}	+0.00(28)	-0.69(28)

High precision ICCs ($\Delta\alpha/\alpha \leq 1.5\%$)

All	All	25	+0.77(51)	-0.56(26)	-0.95(17)
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^{a)} LWM has expanded the uncertainty so the range includes the most precise input value.

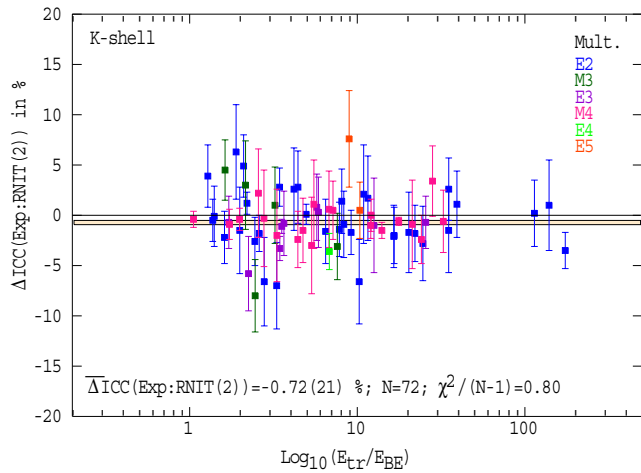


Fig. 4. Same as Fig. 3 but for the "Frozen Orbital", RNIT(2) approximation.

the energy dependence of the $\Delta_{ICC}(Exp:Theor)$ differences to the same scale irrespective of the atomic number. A notable feature of Figure 3 is the correlation between the $E_{tr}/E_{BE} \leq 3$ data points. As is shown in Figure 2, large differences between calculated ICCs are expected close to the shell binding energy. One of the important data points shown is the 80.2369(7) keV pure M4 transition in ^{193}Ir , which is only 4.126 keV above the K-shell binding energy of 76.111 keV. The most accurate experimental value of $\alpha_K(Exp) = 103.0(8)$ reported recently by Nica *et al.* [12] is more than 10 % higher than the $\alpha_K(BTNTR) = 92.18(12)$ obtained with the "No Hole" approximation. When the adopted experimental α_K data set is compared with the theoretical values calculated by the "Self Consistent" and the "Frozen Orbital" approximation slightly smaller average $\Delta_{ICC}(Exp:Theor)$ differences were obtained. For the 80.2369 keV M4 transition the $\alpha_K(RNIT(1)) = 99.63(12)$, $\Delta_{ICC}(Exp:RNIT(1)) = +3.4(8)$ % and $\alpha_K(RNIT(2)) = 103.45(12)$, $\Delta_{ICC}(Exp:RNIT(2)) = -0.4(8)$ %. The ratio of K-shell internal conversion coefficients for the 127.5-keV E3 transition in ^{134}Cs and the 661.7-keV M4 transition in ^{137}Ba determined accurately by Nica *et al.* [14] also prefers the "Frozen Orbital" approximation and disagrees by more than three standard deviations with the "No Hole" calculation.

The main aim of our review was to assemble an experimental data base and to compare it to the various theoretical calculations. The average difference in Table 1 between all experimental data and the new Dirac-Fock model [3] is smaller than 1 %; however, there are some differences in terms of atomic shell and transition multipolarity. It is also evident that the "No Hole" (BTNTR) approximation tends to underestimate experiments, but in some cases $\bar{\Delta}_{ICC}(Exp:BTNTR)$ values show large fluctuations; for example for the α_K and the α_{Total} values for 'All' (see Table 1) and E2 multipolarities. On the other hand the two approximations, "Self Consistent" (RNIT(1)) and "Frozen Orbital" (RNIT(2)) tend to overestimate experiment. However the average differences between the various subgroups in Table 1 do not fluctuate that much. Owing to the fact that almost all transition energies considered were higher than the K-shell binding energy for the decaying

atom, sizable differences in the theoretical ICC values are only expected for the K-shell. It should be noted that the calculated ICC values close to the binding energy of L, M, ... *etc.* atomic shells are also sensitive to the choice of the physical model.

As it is shown in the last entry of Table 1, 25 experimental ICC out of the 186 included in our analysis are known to better than 1.5 % relative uncertainty. The weighted mean values of the $\bar{\Delta}_{ICC}(Exp:Theor)$ differences of this group, comprising 10 α_K , 1 α_L , 5 $\alpha_{K/L}$ and 9 α_{Tot} data, are very similar to the corresponding values obtained for the total data set; however, the uncertainty is factor three larger for "No Hole" (BTNTR) than for "Frozen Orbital" (RNIT(2)). The reduced χ^2 values of BTNTR: 8.2, RNIT(1): 2.12 and RNIT(2): 1.06 (for $N=25$ the critical value at 99 % confidence level is 1.79). This provides a further evidence to support our preference toward the "Frozen Orbital" (RNIT(2)) approximation.

There are other atomic or higher order effects which could influence the ICC values. For example, atomic many body correlations, a non-spherical atomic field in the case of partially filled shells, the uncertainty in the binding energy, the chemical environment of the atom, *etc.* It is assumed that these effects are important close to the shell binding energy. To minimize their impact BrIcc uses an ICC data table starting 1 keV above the shell binding energy.

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