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# Calculation of beta spectral shapes

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**Abstract** – Classical beta spectra calculations are briefly described, highlighting the usual assumptions and limitations. To go beyond these usual assumptions, the numerical resolution of the Dirac equation for the atomic and beta electrons is necessary. This allowed us to determine the parameters  $\lambda_i$  involved in the theoretical shape factors exactly. A systematic comparison between theoretical and experimental shape factors led us to disqualify the usual  $\lambda_i = 1$  assumption for all forbidden transitions. The usual  $\xi$ -approximation was proved to be incorrect for numerous first forbidden non-unique transitions, and for all higherorder non-unique transitions. A more accurate screening correction was defined and the atomic exchange effect was taken into account, an effect which is always neglected in usual calculations. The beta spectra of  $^{63}\text{Ni}$  and  $^{241}\text{Pu}$ , recently measured down to very low energies, are well reproduced in our calculations. The exchange effect was demonstrated to have a great influence on the spectral shape within this energy range.

**Keywords:** radiation, beta / beta particle(s) /  $^{129}\text{I}$  /  $^{58}\text{Co}$  /  $^{99}\text{Tc}$

## 1 Introduction

The study of the shape of beta spectra is experiencing a resurgence of interest having been little studied since the late 1970s. At that time, the knowledge of the spectral shape was thought to be appropriate. However, at present, more precise knowledge of the shape of energy spectra, coupled with well-established uncertainties, is sought by users from the nuclear power industry, the medical care sector and for ionizing radiation metrology.

In ionizing radiation metrology, the energy spectra of beta decays are often needed, especially when measurements are carried out using Liquid Scintillation Counting, since the modeling of the light emission used in the triple to double coincidence ratio (TDCR) method, used to establish a relationship between the detection efficiency and the experimental TDCR ratio, requires the knowledge of the shape of the beta spectra for beta-decaying nuclides (Broda *et al.*, 2007). Due to the threshold effect, the reliability of the TDCR-Cerenkov technique is even more sensitive to the accuracy of the shape of beta spectra (Bobin *et al.*, 2010).

Moreover, biologically targeted radiotherapy using new radionuclides is being developed as a means of cancer treatment, along with the production of new radiopharmaceuticals (Boswell and Brechbiel, 2007). As in any treatment using ionizing radiation, the knowledge of the dose delivered within the tumor, and to the rest of the body, is essential to ensure the efficacy and safety of the treatment. One of the problems is related to the calculation of the dose delivered in the volume of inter-

est according to the quantity of radionuclide which is present there. The precise knowledge of the shapes of beta spectra, particularly in the low-energy part of the spectrum, is essential for calculating the dose deposited at the level of biological cells (Bardiès and Chatal, 1994). Indeed, the very lowenergy electrons have a high contribution to the dose because the linear energy transfer of electrons greatly increases as their kinetic energy decreases (Kassis, 2004).

As the primary laboratory for measurements of radioactivity in France, the LNHB requires a computer code, which should be validated against experiments. Hence, the evaluation of the beta spectral shapes leads us to be interested in both experimental and theoretical studies. This contribution is focused on the latter. First, the classical beta spectra calculations are described, highlighting the chosen assumptions and their limitations. Then, our current theoretical abilities will be summarized in order to illustrate how it is possible to go beyond these usual assumptions.

## 2 Usual beta spectra calculations

During the beta decay of a radionuclide, the available energy is split between the electron, the neutrino and the daughter nucleus. This three-body decay implies a continuous energy spectrum for electrons. The shape of this spectrum is influenced by the nature of the transition that can be “allowed” or “forbidden” depending on the structure of the nuclei involved, and by certain atomic effects. Electrons thus have continuous energy spectra of varied shapes.

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**Table 1.** Classification of the nature of beta transitions according to the variations of the total angular momentum ( $\Delta J$ ) and parity ( $\Delta\pi$ ) of the initial and final nuclear states.

$ \Delta J $	$\Delta\pi$	Nature of transition
0, 1		Allowed
0, 1	-1	1st forbidden non-unique
> 1	$(-1)^{ \Delta J }$	$ \Delta J $ th forbidden non-unique
> 1	$(-1)^{ \Delta J -1}$	$( \Delta J  - 1)$ th forbidden unique

## 2.1 Basic formulation

A beta spectrum is described by a weak interaction coupling constant,  $g$ , a statistical phase space factor, the so-called Fermi function  $F$  which corrects for the Coulomb effects, and a shape factor,  $C$ . The phase space factor simply reflects the momentum distribution between the electron and the neutrino. The shape factor contains all of the remaining energy dependencies, such as leptonic and nuclear matrix elements or corrections for atomic effects. Thus, the number of beta particles emitted per unit of time with the energy  $W$ , in the range between  $W$  and  $W + dW$  is (Behrens and Jänecke, 1969):

$$N(W) dW = \frac{g^2}{2\pi^3} p W (W_0 - W)^2 F(Z, W) C(W) dW \quad (1)$$

where: the normalized energy is  $W = 1 + E_{kin}/m$ , with  $E_{kin}$  the kinetic energy of the electron and  $m$  its mass at rest; the corresponding normalized momentum is  $p = \sqrt{W^2 - 1}$ ; the end-point normalized energy is  $W_0 = 1 + E_{max}/m$ , with  $E_{max}$  the end-point energy of the electro; and  $Z$  is the atomic number of the daughter nucleus.

The nature of the beta transition depends on the variations of the total angular momentum ( $\Delta J$ ) and parity ( $\Delta\pi$ ) of the initial and final nuclear states. The corresponding classification is given in Table 1.

## 2.2 Theoretical shape factors

The shape factor depends on the nature of the transition and on the nuclear matrix elements of the weak interaction operators, but their exact calculation is a difficult task. In the case of allowed and unique forbidden transitions, the energy dependence of the nuclear matrix elements can be factored out (Gove and Martin, 1971). Thus, their energy independent part just appears as a normalization factor in the calculation of the shape of a given beta spectrum, and can be ignored. This factorization is only possible because of the assumption that the contribution of the leptonic wave functions over the entire space can be replaced by their value at the nuclear surface. Then, the shape factors for allowed and forbidden unique transitions can be established (Behrens and Bühring, 1982). They are summarized in Table 2 up to a third forbidden unique transition, with the neutrino momentum defined as  $q = W_0 - W$ .

The  $\lambda_i$  parameters are the Coulomb amplitudes of the electron wave functions. They have to be calculated numerically and the procedure is not straightforward. For this reason, they are often set to 1 in classical beta spectra calculations, even in the most recent ones (Huber, 2011).

**Table 2.** Theoretical shape factors up to a third forbidden unique transition. See text for the explanation of the parameters.

Nature of the transition	Shape factor $C(W)$
Allowed	1
1st forbidden unique	$q^2 + \lambda_2 p^2$
2nd forbidden unique	$q^4 + (10/3)\lambda_2 q^2 p^2 + \lambda_3 p^4$
3rd forbidden unique	$q^6 + 7\lambda_2 q^4 p^2 + 7\lambda_3 q^2 p^4 + \lambda_4 p^6$

In forbidden non-unique transitions, the energy dependence of the nuclear matrix elements cannot be factored out, which greatly complicates the calculation of the beta spectral shapes. However, first forbidden non-unique transitions which fulfil the following assumption can be treated as allowed ones (Schopper, 1966):

$$2\xi = \alpha Z/R \gg E_{max} \quad (2)$$

with  $\alpha$  the fine structure constant and  $R$  the radius of the daughter nucleus. This approximation, also called the  $\xi$ -approximation, simply means that the Coulomb energy of the beta electron at the nuclear surface must be large compared with the total decay energy. Typically, this approximation is applied to all forbidden non-unique transitions, calculating each as if it were a unique transition with the same variation of the total angular momentum. There is no theoretical justification for this generalization.

## 2.3 Atomic screening effect

When calculating a beta spectrum, the atomic screening effect is generally corrected by using a constant Thomas-Fermi potential which is subtracted from the total energy of the particle (Good, 1954). This method has the advantage of being analytical, but it creates a non-physical discontinuity at the minimum energy defined by the potential (Mougeot *et al.*, 2011). Physically the influence of the atomic electrons is expected to be significant when the beta wavelength is comparable with the size of the atom at low energy.

## 2.4 Other effects

Two other effects are usually taken into account, namely the finite nuclear size effect (Gove and Martin, 1971) and the radiative corrections (Behrens and Bühring, 1982); mainly because analytical formulae exist. More details can be found in (Mougeot *et al.*, 2011).

A computer code, BetaShape, has been developed at the LNHB to calculate the beta spectra for allowed and forbidden unique transitions according to the assumptions described in this section (Mougeot *et al.*, 2011). The  $\lambda_i$  parameters used therein are determined following the method given in (Gove and Martin, 1971). BetaShape has been developed as a C++ class, which was successfully implemented in our Geant4 simulation code.

### 3 How to go beyond the usual assumptions

As has been shown in (Mougeot *et al.*, 2012), these usual assumptions, which allow analytical wave functions to be used, are not reasonable for high  $Z$  and at low energies. In these cases, atomic effects have to be correctly taken into account. Moreover, it is also necessary to know if the  $\xi$ -approximation can be reasonably applied.

#### 3.1 Relativistic electron wave functions

As spin-half particles, electrons follow the Dirac equation. Atomic electrons are in bound states, while beta electrons are in continuum states. Implicit in the assumptions of Section 2 is that the Coulomb potential is generated by a pointcharge nucleus.

We adopted a better approach, by considering the nucleus to be a uniformly charged sphere with a correction for the influence of the atomic electrons. Such screened potentials can be found in (Salvat *et al.*, 1987). However, the Dirac equations for the wave functions can no longer be solved analytically. Hence, we followed the numerical method described in detail in (Behrens and Bühring, 1982).

The electron radial wave functions are expressed locally as power series expansions, allowing the Dirac equations to be solved using recurrence relations. Special treatment is needed for the solutions relative to the regular singular point  $r = 0$  and the irregular singular point  $r = \infty$ . Then, the procedure consists of evaluating the wave functions at the two limits, *i.e.* near  $r = 0$ , and at a point sufficiently far away from the nucleus for a correct evaluation with the expansion near  $r = \infty$ , followed by stepbystep calculations between these two points in order to reconnect each solution with the appropriate renormalization and phase shift.

In order to be sure of our calculations, the tables from (Behrens and Jänecke, 1969) of various parameters used to calculate beta spectra were recalculated. Despite a lack of precise and detailed information on the exact way these parameters had been calculated, the unscreened Coulomb functions, Table II in (Behrens and Jänecke, 1969), were well reproduced, and the ratios of screened to unscreened Coulomb functions, Table III in (Behrens, 1969), were in very good agreement for all  $Z$ . We also obtained good agreement with the parameters of Table V in (Behrens and Jänecke, 1969) for the bound wave functions.

#### 3.2 Theoretical shape factors

These precise electron wave functions allow us to calculate the  $\lambda_i$  parameters exactly at any desired energy. It is therefore possible to test the assumption  $\lambda_i = 1$  and the generalization of the  $\xi$ -approximation.

We calculated 130 beta transitions for which we could find an experimental shape factor. These shape factors were mostly taken from (Behrens and Szybisz, 1976). We tested 36 allowed transitions, 25 first forbidden unique, 4 second forbidden unique, 1 third forbidden unique, 53 first forbidden non-unique, 9 second forbidden non-unique, 1 third forbidden non-unique and 1 fourth forbidden non-unique.

As expected, there is good agreement between theoretical and experimental shape factors for allowed and forbidden unique transitions when the  $\lambda_i$  are correctly calculated. The  $\lambda_i = 1$  assumption leads to a systematically worse agreement, modifying the mean energy of the beta spectrum by several percent.

The  $\xi$ -approximation appears to be valid for about half of the tested first forbidden non-unique transitions. This is consistent with the limitations given in (Schopper, 1966). For all other cases, and especially for higherorder non-unique transitions, this approximation is clearly inappropriate, modifying the mean energy of the beta spectrum by several tens of percent. Some tested beta transitions are given in Table 3 to illustrate this.

#### 3.3 Atomic effects

At low energy, atomic effects play an important role in determining the shape of beta spectra. The sudden change in the nuclear charge can induce atomic excitations or internal ionizations, respectively called *shake-up* and *shake-off* effects, but these are not expected to have a contribution higher than 0.1%. The two major atomic effects are screening and the exchange effect.

##### 3.3.1 Screening

Atomic electrons partially screen the nuclear charge seen by the beta electron. As seen in Section 2, beta spectra are classically calculated using the electron radial wave functions evaluated at the nuclear surface. However, to take the screening into account, it is not sufficient to just renormalize the wave functions and evaluate them at the nuclear radius, because the screened potentials are very weak at this distance, and so the modification is completely negligible over the entire range of the spectrum. In fact, it is necessary to take into account the spatial extension of the wave functions. As described in (Behrens and Bühring, 1982), this can be done by evaluating the transition matrix of the corresponding beta decay. However the calculations become very complicated because the electron wave functions are coupled with the nuclear matrix elements involved. To avoid this difficulty, we have defined a new, simple type of screening correction available for allowed transitions (Mougeot *et al.*, 2014).

##### 3.3.2 Exchange effect

The exchange effect is also an atomic effect. It arises from the creation of a beta electron in a bound orbital of the daughter atom corresponding to one which was occupied in the parent atom. An atomic electron from the bound orbital simultaneously makes a transition to a continuum orbital of the daughter atom. This process cannot be distinguished from the direct decay to a final state containing one continuum electron.

A formalism for the exchange effect has already been set out in detail in (Harston and Pyper, 1992) for allowed transitions. Indeed in this case, only the  $ns$  orbitals are reachable by the beta electron, which greatly simplifies the calculations.

**Table 3.** Experimental mean energies and differences from calculated mean energies for a set of second (*2nuf*), third (*3nuf*) and fourth (*4nuf*) forbidden non-unique transitions. Two calculations are performed: setting  $\lambda_i = 1$  or calculating these parameters exactly (*BS*). The nature of the transition used according to the  $\xi$ -approximation is also given: first (*1uf*), second (*2uf*) and third (*3uf*) forbidden unique.

Nucleus	Nature	$\xi$ -approximation	$E_{\max}$ (keV)	$E_{\text{moy}}$ exp (keV)	$\Delta E_{\text{moy}}$ BS (%)	$\Delta E_{\text{moy}}$ $\lambda_i = 1$ (%)
$^{36}\text{Cl}$ ( $\beta^-$ )	2nuf	as 1uf	709.53	314.09	-11.47	-11.02
		best as 3uf			3.51	4.26
$^{46}\text{Sc}$ ( $\beta^-$ )	2nuf	as 1uf	1477.4	657.83	-8.40	-7.90
		best as 3uf			-0.79	-0.10
$^{59}\text{Fe}$ ( $\beta^-$ )	2nuf	as 1uf	1565.2	583.00	8.61	9.47
		best as allowed			5.00	5.00
$^{58}\text{Co}$ ( $\beta^+$ )	2nuf	as 1uf	1285.4	708.42	-17.73	-17.20
		best as 3uf			-11.58	-10.92
$^{94}\text{Nb}$ ( $\beta^-$ )	2nuf	as 1uf	471.5	94.38	75.57	81.77
		best as allowed			54.10	54.10
$^{99}\text{Tc}$ ( $\beta^-$ )	2nuf	as 1uf	293.8	95.28	5.88	11.68
		best as allowed			-11.28	-11.28
$^{129}\text{I}$ ( $\beta^-$ )	2nuf	as 1uf	151.2	36.09	35.23	53.87
		best as allowed			12.36	12.36
$^{135}\text{Cs}$ ( $\beta^-$ )	2nuf	as 1uf	268.7	61.57	44.15	56.55
		best as allowed			22.76	22.76
$^{137}\text{Cs}$ ( $\beta^-$ )	2nuf	as 1uf	1175.63	284.90	48.85	54.41
		best as allowed			45.35	45.35
$^{87}\text{Rb}$ ( $\beta^-$ )	3nuf	as 2uf	283.3	56.65	101.7	115.1
		best as allowed			44.6	44.6
$^{113}\text{Cd}$ ( $\beta^-$ )	4nuf	as 3uf (best)	322	139.83	-1.08	8.61

The exchange effect is determined in our calculations following this formalism and using the bound and continuum electron wave functions described in Section 3.1.

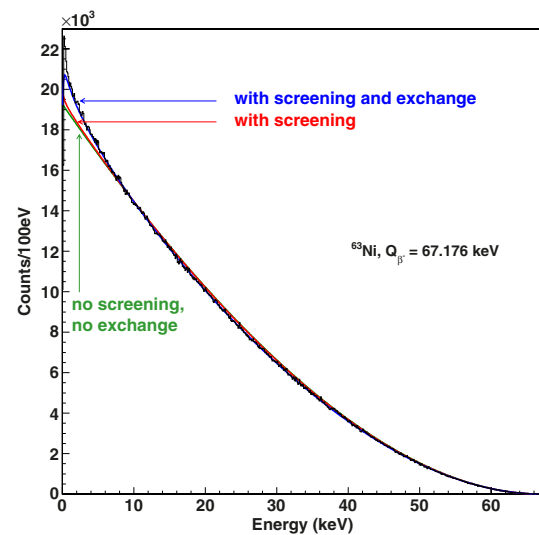
## 4 Results

Accurate calculations, based on inclusion of the various effects covered above, were performed for the allowed transition of  $^{63}\text{Ni}$  and the first forbidden non-unique transition of  $^{241}\text{Pu}$ .

$^{63}\text{Ni}$  decays entirely by beta minus emission to the ground state of  $^{63}\text{Cu}$ . This allowed transition has a maximum energy of 66.98 keV (Bé *et al.*, 2006).

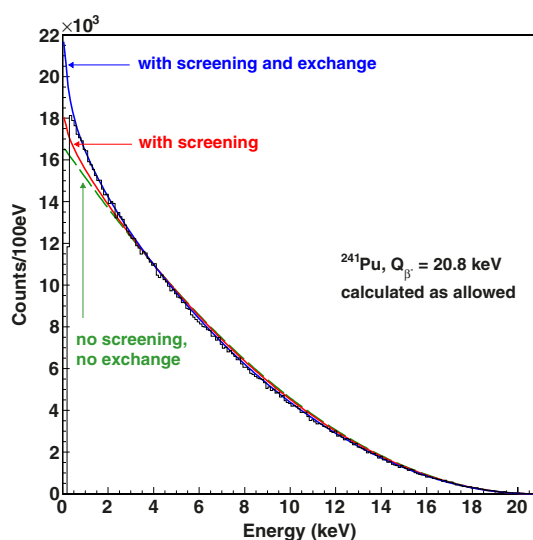
$^{241}\text{Pu}$  decays mainly by beta minus emission to the ground state of  $^{241}\text{Am}$ . This first forbidden non-unique transition has a maximum energy of 20.8 keV (Bé *et al.*, 2008). This transition fulfills the assumption of the  $\xi$ -approximation (see Sect 2.2) and can be calculated as allowed (Schopper, 1966).

The low maximum energies of these two transitions make them ideal cases for evaluating the influence of the atomic effects. The beta spectra of  $^{63}\text{Ni}$  (LeBret, 2012) and  $^{241}\text{Pu}$  (Loidl *et al.*, 2010) were recently measured using metallic magnetic calorimeters. Each source was enclosed in a gold absorber assuring a  $4\pi$  solid angle and 100% detection efficiency. These spectra are first compared with classical beta calculations, and then taking into account the screening and exchange effects. The results of our calculations are compared with the measured spectra in Figure 1 for  $^{63}\text{Ni}$  decay and in Figure 2 for  $^{241}\text{Pu}$  decay.



**Fig. 1.** Comparison between the measured beta spectrum of  $^{63}\text{Ni}$  from (LeBret, 2012) and three calculated spectra: a classical allowed transition without screening or exchange, the same including screening, and a complete one including screening and exchange corrections.

Excellent agreement was obtained in both decays over the entire energy range of the spectrum, which validates our calculations and the influence of the atomic effects on beta spectral shapes. For  $^{63}\text{Ni}$ , taking the exchange effect into account leads to a mean energy which is lower by about 1.8%. For  $^{241}\text{Pu}$ , the mean energy of the beta spectrum is lower by about



**Fig. 2.** Comparison between the measured beta spectrum of  $^{241}\text{Pu}$  from (Loidl, 2010) and three calculated spectra: a classical allowed transition without screening or exchange, the same including screening, and a complete one including screening and exchange corrections.

4% when both the screening and exchange effects are included in the calculations. As expected, the influence of screening is weak for the low- $Z$  nuclide  $^{63}\text{Ni}$  but has a significant contribution, equivalent to the magnitude of the exchange effect, for the high- $Z$  nuclide  $^{241}\text{Pu}$ .

## 5 Conclusion

Classical beta spectra calculations have been described, highlighting the assumptions and their limitations. To go beyond these usual assumptions, the numerical resolution of the Dirac equations is necessary for both the atomic electrons and the beta electrons. Using this method, we were able to determine the  $\lambda_i$  parameters involved in the theoretical shape factors exactly.

A systematic comparison between theoretical and experimental shape factors leads us to disqualify the  $\lambda_i = 1$  assumption for all forbidden transitions. The  $\xi$ -approximation was also proved not to be correct for numerous first forbidden non-unique transitions, and for all higherorder non-unique transitions.

Our determination of the screening effect is more accurate than the usual one, because we take into account the spatial extension of the electron wave functions. For the first time, we took into account the atomic exchange effect, which has been systematically ignored in all previous calculations. The exchange effect was demonstrated to have a great influence on the spectral shape at low energy. The shape of the beta spectra of  $^{63}\text{Ni}$  and  $^{241}\text{Pu}$  are reproduced well in our calculations, even at very low energies.

We now plan to evaluate the influence of the nuclear matrix elements in order to calculate specifically the forbidden

non-unique transitions. Both the screening and exchange effects will have to be taken into account exactly. These calculations will be compared with new measurements carried out at the LNHB.

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