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Abstract

In ionizing radiation metrology, the energy spectra of beta decays are often needed, especially when measurements are carried out using Liquid Scintillation Counting. The uncertainties on activity measurements can be reduced with a precise knowledge of the shapes of the beta spectra. The few studies which have been conducted on this subject since the 1970s were mainly focused on allowed and first non-unique forbidden transitions. But it is also necessary to have a good knowledge of spectra for unique and non-unique forbidden transitions of higher orders. Our goal is to develop a computer program for calculating beta spectra, validated by experiments, including evaluated uncertainties on the shapes. The exhaustive evaluation of form factors measured so far shows that there are not enough good data to validate those complex calculations. All these considerations highlight the need for a consistent experimental program in which all the distortion phenomena of beta spectra should be analysed and quantified very carefully. This paper describes the present features of the computer program we have developed, BetaShape, and specifies the future work which should be undertaken to improve it.

1. Introduction

As evaluators of nuclear decay data, we notice an increasing demand from users for a precise knowledge of the shape in energy of beta spectra, coupled with well established uncertainties. For example, these spectra are useful in nuclear power plants for calculations of the residual power, or in medical care for calculations of doses delivered to patients' cells. In ionizing radiation metrology, a better knowledge of the spectra would reduce the uncertainties on the activity measurements carried out by ionisation chambers or Liquid Scintillation Counting.

To answer the users' demand, calculations are needed because all the wanted beta spectra cannot be measured: very short half-lives, compound spectra, etc. Carrying out experiments is also necessary to test and validate the models. The critical point is then to quantify all the phenomena that distort a measured beta spectrum: backscattering, bremsstrahlung, response function of the detector, dead zones, source quality (thickness, size and impurities), etc. Thanks to the increasing power of the computers, it is now possible to consider more complex models for the theory, and more complex Monte-Carlo simulations for the experiments.

While the allowed transitions are well known, this is not the case for spectra of unique and non-unique forbidden transitions of higher orders. V. Gorozhankin made a bibliographical compilation of the published form factors up to 2003 [11Go] in order to determine the mean form factors which could be applied to all spectra. However, it is clear that there are not enough experiments to validate the theoretical evaluations of forbidden transitions. Only about ten form factors have been published since 1976, and concerning the theory we are in the same situation: theorists are not working anymore on beta spectra.

The analysis of various available computer codes led us to develop our own program, which is called BetaShape. BetaShape was developed as a C++ class, allowing an easy

implementation in a Monte-Carlo simulation code or in a nuclear database program. Most of these codes calculate only beta spectra of allowed transitions, and give the possibility to add an experimental shape factor. The energy dependence of these shape factors is usually determined using a Kurie plot (defined in section 2.5). The energy parameterizations are summarized in [76Be] and [11Go]. The Radlist program [88Bu] calculates spectra up to the third unique forbidding order without experimental shape factor. We have decided to follow the same approach and to improve it.

2. Calculation method

2.1 Basic formulae

A beta spectrum is the result of a weak interaction coupling constant *g*, a statistical phase space factor which reflects the momentum distribution between the electron and the neutrino, the so-called Fermi function *F* which corrects for the Coulomb effects, and a shape factor *C* which contains the relevant information about the nuclear structure. The number of beta particles emitted per unit of time with the energy *W*, in the range between *W* and *W*+*dW*, is [69Be]:

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

where: the normalized energy $W = 1 + E_{kin}/m$, with E_{kin} the kinetic energy of the particle and *m* the electron mass at rest; the corresponding normalized momentum $\eta = (W^2\text{-}1)^{1/2}$; the end-point normalized energy $W_0 = 1 + E_{max}/m$, with E_{max} the end-point energy of the particle; and Z the atomic number of the daughter nucleus.

The transition order depends on the variations of the total angular momentum (ΔJ) and of the parity of the initial and final nuclear states. The shape factor *C*(*W*) depends on the transition order 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 [71Go]. 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 left out. This factorization is not possible in the case of the non unique forbidden transitions. The specific calculation of these transitions is difficult because of the large number of nuclear matrix elements to calculate. A usual approximation is to calculate non-unique transitions as unique transitions having the same variation of the total angular momentum.

As in the Radlist program [88Bu], the BetaShape program follows the Gove and Martin's approach [71Go]. The calculated spectrum is:

$$N(W) = \eta W (W_0 - W)^2 S_n(Z,W) \quad (2)$$

with:

$$S_n(Z,W) = \sum_{k=1}^{n+1} \frac{\lambda_k(Z,W)\eta^{2(k-1)}(W_0 - W)^{2(n-k+1)}}{(2k-1)![2(n-k+1)+1]!} \quad (3),$$

$$\lambda_k(Z,W) = \frac{g_{-k}^2 + f_{+k}^2}{2\eta^2}\left[\frac{(2k-1)!!}{(\eta R)^{k-1}}\right]^2 \quad (4),$$

and $R = 0{,}002908A^{1/3} - 0{,}002437A^{-1/3}$ the radius of the nucleus in $\hbar/mc$ units. The parameter n equals $|\Delta J|-1$, with a value of zero for the allowed and first non unique forbidden transitions. $f_\kappa(r)$ and $g_\kappa(r)$ are the electronic radial wave functions, continuum state solutions of the Dirac equation for an electron into a Coulomb field [82Be]:

$$\begin{aligned} \frac{d}{dr}g_\kappa(r) + \frac{\kappa+1}{r}g_\kappa(r) - \left[E_{tot} + m - V(r)\right]f_\kappa(r) = 0 \\ \frac{d}{dr}f_\kappa(r) - \frac{\kappa-1}{r}f_\kappa(r) + \left[E_{tot} - m - V(r)\right]g_\kappa(r) = 0 \end{aligned} \quad (5)$$

where E_{tot} is the total particle energy, $V(r)$ the Coulomb potential generated by the Z protons of the nucleus, and κ the angular momentum quantum number. κ, a non zero relative integer, is the eigenvalue of the operator $K = \beta\left[\vec{\sigma}\cdot\vec{L} + 1\right]$, with β the Dirac's matrix, $\vec{\sigma}$ the Pauli's matrices and $\vec{L} = \vec{r}\times\vec{p}$ the classical angular momentum operator.

If the nucleus is considered as a point source, the equations (5) can be solved analytically and the calculations are fast. They can also be solved numerically: the description of the nucleus can be then more realistic, but the calculations need a lot of computing power. Therefore, theorists used to generate tables of data from which it is necessary to interpolate some values in order to calculate the needed beta spectra (Behrens tables [69Be], Dzhelepov tables [69Dz]).

N.B. Gove and M.J. Martin used the results from M.E. Rose for the expression of the electronic radial wave functions [61Ro]. These wave functions, calculated analytically in a central Coulomb potential, are:

$$\begin{aligned} f_k &= \sqrt{W-1}\,Q_k\,\Im[S_{k\,1}F_1(a,b,z)] \\ g_k &= \sqrt{W+1}\,Q_k\,\Re[S_{k\,1}F_1(a,b,z)] \end{aligned} \quad (6)$$

with:

$$Q_k = \frac{(2\eta R)^{\gamma_k}\, e^{\pi y/2}\left|\Gamma(\gamma_k + iy)\right|}{2R\sqrt{W}\,\Gamma(2\gamma_k+1)} \quad (7), \qquad S_k = (\gamma_k + iy)e^{-i\eta R}\sqrt{\frac{-k+iy/W}{\gamma_k+iy}} \quad (8),$$

$$\gamma_k = \sqrt{k^2-(\alpha Z)^2}\,,\ y = \frac{\alpha ZW}{\eta}\,,\ a = (\gamma_k+1)+iy\,,\ b = (2\gamma_k+1)\,, \text{ and } z = 2i\eta R\,. \quad (9)$$

Conventionally, *Z* is positive for beta minus decays and negative for beta plus decays. ${}_1F_1(a,b,z)$ is the confluent hypergeometric function, defined using the $\Gamma(z)$ function. α is the fine structure constant.

When the particle momentum tends to zero, the electron spectrum tends to a finite value while the positron spectrum tends to zero. For *W* less than $1+5\cdot10^{-6}$, the calculation of the electron wave functions is thus modified:

$$\begin{Bmatrix} f_k \\ g_k \end{Bmatrix}(\beta^-) = \frac{2\sqrt{2\pi\eta}\,(2R)^{\gamma_k-1}(\alpha Z)^{\gamma_k-1/2}}{\Gamma(2\gamma_k+1)} \begin{Bmatrix} \dfrac{(\alpha Z)^2}{2}\left(1-\dfrac{4\alpha ZR}{2\gamma_k+1}\right) \\ \left[-k(\gamma_k-k)-\dfrac{(\alpha Z)^2}{2}\right]\left(1-\dfrac{4\alpha ZR}{2\gamma_k+1}\right)-\dfrac{2\alpha ZR(\gamma_k-k)}{2\gamma_k+1} \end{Bmatrix}^{1/2} \quad (10)$$

Three correction terms are added: the screening effect due to the electron cloud of the daughter atom, the finite nuclear size effect with regard to the electron Compton wavelength, and the radiative corrections.

2.2 Screening effect correction

To correct for the screening effect, the method of R.H. Good Jr is used [54Go], taking a potential V_0 which depends on the beta decay type:

- $V_0(\beta^-) = 6{,}971217 \cdot 10^{-5} Z^{1{,}378983}$, and W becomes (W-V_0) if (W-V_0)>1,
- $V_0(\beta^+) = V_0(\beta^-) \exp\left(\frac{a}{\eta} + \frac{b}{\eta^2}\right)$, (11)

$$a = 1{,}11 \cdot 10^{-7} Z^3 - 1{,}01 \cdot 10^{-5} Z^2 - 2{,}38 \cdot 10^{-3} Z + 0{,}102,$$

$$b = -2{,}42 \cdot 10^{-8} Z^3 + 3{,}83 \cdot 10^{-6} Z^2 + 3{,}60 \cdot 10^{-5} Z - 0{,}0156,$$

and W becomes $W+V_0$.

For beta minus transitions, this correction is applied only above a threshold energy mV_0, which induces a non-physical discontinuity in the spectrum. For Z less than 100, this threshold is less than 20 keV. The normalized momentum η is recalculated with the new W value. These new values are introduced in the calculation of $S_n(Z,W)$ and in the phase space factor, except in the neutrino energy (W_0-W).

2.3 Finite nuclear size correction

The finite nuclear size effect reflects the influence of the nuclear charge distribution on the beta spectrum. The adopted correction is given by [71Go]:

- $\lambda_k(Z,W)$ becomes $\lambda_k \cdot [1 + \Delta\lambda_k]$,
- $\Delta\lambda_k = 0$ if $k > 1$, or $Z(\beta^-) \le 50$, or $Z(\beta^+) \le 80$,
- $\Delta\lambda_1(\beta^-) = (Z-50)[-25 \cdot 10^{-4} - 4 \cdot 10^{-6}(Z-50)W]$ if $Z(\beta^-) > 50$, (12)
- $\Delta\lambda_1(\beta^+) = (Z-80)\left[-17 \cdot 10^{-5} W + \frac{63 \cdot 10^{-5}}{W} - \frac{88 \cdot 10^{-4}}{W^2}\right]$ if $Z(\beta^+) > 80$.

This correction is applied for heavy nuclei and if k = 1 only. The authors in [71Go]

determined it from the $\Delta\lambda_k(Z,W)$ plots published by M.E. Rose and D.K. Holmes ([51Ro], [57Ro]).

2.4 Radiative corrections

The radiative corrections take into account the energy loss of the electron in the electromagnetic field of the nucleus (virtual photons and internal bremsstrahlung). They are calculated as described in [82Be], [67Si], and [72Ja]. The spectrum is corrected for using a factor $[1+\delta_R(W,Z)]$ calculated as follows:

$$\delta_R(W,Z) = \delta_1(W) + \delta_2(Z) + \delta_3(Z) + \delta_4(Z) \quad (13)$$

with:

- $\delta_1(W) = \frac{\alpha}{2\pi} g(W,W_0)$,

  $$g(W,W_0) = 3\ln\left(\frac{m_p}{m}\right) - \frac{3}{4} + 4\left(\frac{arcth\beta}{\beta} - 1\right)\left\{\frac{W_0 - W}{3W} - \frac{3}{2} + \ln\left[2(W_0 - W)\right]\right\} + \frac{4}{\beta} L\left(\frac{2\beta}{1+\beta}\right) + \frac{arcth\beta}{\beta}\left[2(1+\beta^2) + \frac{(W_0 - W)^2}{6W^2} - 4arcth\beta\right] \quad (14),$$

  $\beta = \eta / W$, $L(x) = \int_0^x \frac{\ln(1-t)}{t} dt$ the Spence function, and m_p the proton mass at rest,

- $\delta_2(Z) = 1.1|Z|\alpha^2 \ln\left(\frac{m_p}{m}\right)$, (15)

- $\delta_3(Z) = \frac{Z^2\alpha^3}{\pi}\left(3\ln 2 - \frac{3}{2} + \frac{\pi^2}{3}\right)\ln\left(\frac{m_p}{m}\right)$ (16),

- $\delta_4(Z) = \frac{|Z|\alpha^3}{2\pi}\ln\left(\frac{2m_p}{m}\right)$. (17)

2.5 Remarks

The beta spectrum is evaluated using equation (2) for a large number of energies. It is then normalized by its integral and the mean energy is determined. If an experimental spectrum is available, the BetaShape program calculates also the corresponding Kurie plot defined by:

$$K(W) = \sqrt{\frac{N_{\exp}(W)}{\eta W S_n(Z,W)}}. \quad (18)$$

3. Comparison of the results

3.1 Radlist

We compared the results of our calculations with those of the Radlist program [88Bu], which uses the same assumptions but does not take into account the radiative corrections. Thus, we switched them off for these comparisons. Numerical integration methods and digit precisions (double in BetaShape, float in Radlist) are also different.

We considered a wide range of *Z*, *A*, end-point energies and transition orders. A total of 38 transitions, listed in Table 1, were calculated. The non-unique transitions are calculated as unique transitions having the same variation of the total angular momentum.

Both codes give the same results over almost the entire spectrum (discrepancy less than 1 %). Because of the difference of the digit precisions, the energy thresholds mV_0 of the screening correction present a difference of about 200 eV. This implies a difference between 3 and 5 % over a narrow range, typically in one energy bin. For beta plus decay, the screening correction depends on the particle energy, and thus induces a shift over the entire spectrum. A difference higher than 10 % could be observed at very low energies, where this contribution has the greatest influence.

3.2 Beta Spectrum

The Beta Spectrum program has been developed by V. Gorozhankin *et al.* [11Go]. This program calculates the beta spectra shapes using the Behrens tables or the Dzhelepov tables for the Fermi function, according to the user's choice.

The Behrens and Dzhelepov calculations correct for the screening effect and the finite nuclear size effect, but do not take into account the radiative corrections. The Dzhelepov tables are given from 5 keV to 10 MeV. The Behrens tables cover the range 2.6 keV to 25 MeV without corrections, so it is necessary to correct these data using other tables, also provided by Behrens, but more restricted in energy. V. Gorozhankin *et al.* showed that the

choice of the tables used does not induce discrepancies higher than 1 % in most cases. Discrepancies around 5 % could occur for radionuclei having a low *Z* or an end-point energy lower than 100 keV.

We compared the results of our calculations with those obtained with Beta Spectrum for the allowed and first non unique forbidden transitions listed in Table 1. We switched off the radiative corrections and we used the Dzhelepov tables. If we compare the relative difference between the two programs, the discrepancy is about 0.5 % and some fluctuations are present, which mainly come from the interpolation between the tabulated values of the Fermi function. It is noteworthy that this good agreement is obtained by switching off the screening correction in BetaShape. Otherwise, the discrepancy can be higher than 1 % in some cases, suggesting that the screening effect was not corrected in the Dzhelepov tables.

4. Influence of the various corrections

The magnitude of each correction is detailed in Table 2 for some nuclei.

In the calculation model of the BetaShape program, the spectrum shape depends on the adopted transition order. To illustrate the influence of the transition order on the spectra calculated by BetaShape, we took the ^{99}Tc spectrum as an example. Four cases are considered, as described in Figure 1: an allowed, first, second and third unique forbidden transitions. The shapes are very different from the beginning to the end of the spectrum and the mean energies differ by at least 15 %. The true order of this transition is second non-unique forbidden, calculated as a first unique forbidden according to our assumptions. Obviously this approximation needs an experimental validation, with a careful analysis of the distortion sources of the measured spectrum.

One can observe, in Figure 1, a discontinuity at low energy due to the screening correction. As described in Section 2, it occurs below 20 keV for *Z* less than 100. This discontinuity could easily be smoothed out using different kinds of functions, but each

function will affect the spectrum shape differently. Thus, we need some experimental spectra to derive a smooth function. The radiative corrections and the finite nuclear size correction are usually weak. The finite nuclear size correction is less than 0.5 %. The radiative corrections are important near the end-point energy (several tenths of percents), but as the statistics of the spectrum is low in this range, their influence on the spectrum shape is limited.

5. Experimental spectrum of ^{241}Pu

For spectra having low end-point energy, the metallic magnetic calorimeters will be helpful to obtain high precision measurements. The beta spectrum of ^{241}Pu was measured at LNHB with a metallic magnetic calorimeter, as described elsewhere [10Lo]. It can be observed in Figure 2, with the X-rays which come from the decay of a ^{55}Fe source used for the energy calibration. The corresponding Kurie plot is given in Figure 3.

The ^{241}Pu beta decay is a first non-unique forbidden transition, calculated as an allowed one. Our calculation is in good agreement with the experimental results, except at low energy. All the sources of distortion are not yet completely understood, but this discrepancy does not seem to come from the measured spectrum. We are currently studying the possibility of adding a correction to take into account the exchange effect. This effect corresponds to the bound solutions of the coupled differential equations (1). The initial and final state orbitals are usually non orthogonal, allowing an electron to be created in an occupied orbital of the daughter atom. The electron, previously occupying the atomic orbital, is ejected with an energy corresponding to the transferred momentum. According to M.R. Harston and N.C. Pyper [92Ha], the exchange effect for ^{241}Pu could distort the beta spectrum by increasing the channel contents by about 10 % below 2 keV.

6. Conclusion

In order to answer users' demand, we have developed a specific program dedicated to the calculation of beta spectra shapes. BetaShape is based on the analytical approach from Gove and Martin described above. The Coulomb corrections depend on the forbidding order of the transition, but the non-unique forbidden transitions are not yet treated specifically. The screening effect due to the electron cloud, the finite nuclear size effect with regard to the electron Compton wavelength, and the radiative corrections are taken into account. This program must be tested and validated by experiments.

There is a lack of data to test the transitions with high forbidding order. Moreover, the oldest published experiments did not have the computing power for Monte-Carlo simulations to evaluate all the physical phenomena that could distort the spectra. It is necessary to carry out new and well analysed experiments. Metallic magnetic calorimeters provide very encouraging results. The analysis of the ^{241}Pu beta spectrum leads us to take into account the exchange effects in the calculations (currently in progress). Also, we are working on another experimental setup using a silicon detector. This setup will be complementary to the metallic magnetic calorimeters, providing spectra with energy ranging from 100 keV to 1 MeV.

Acknowledgment

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Figures

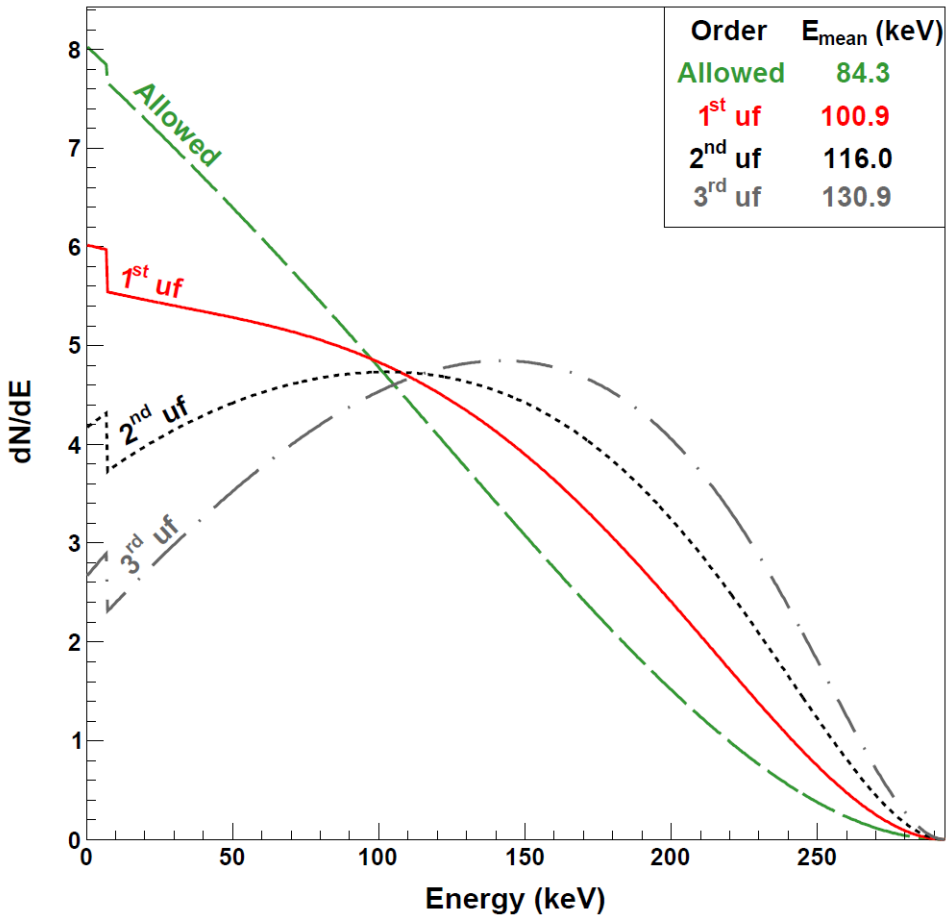

Figure 1 – ^{99}Tc is a second non unique forbidden transition, with an end-point energy of 293.6 keV. The present beta spectra were calculated with different forbidding orders (*uf* means *unique forbidden*). The shapes, mean energies and most probable energies are very different from one to another.

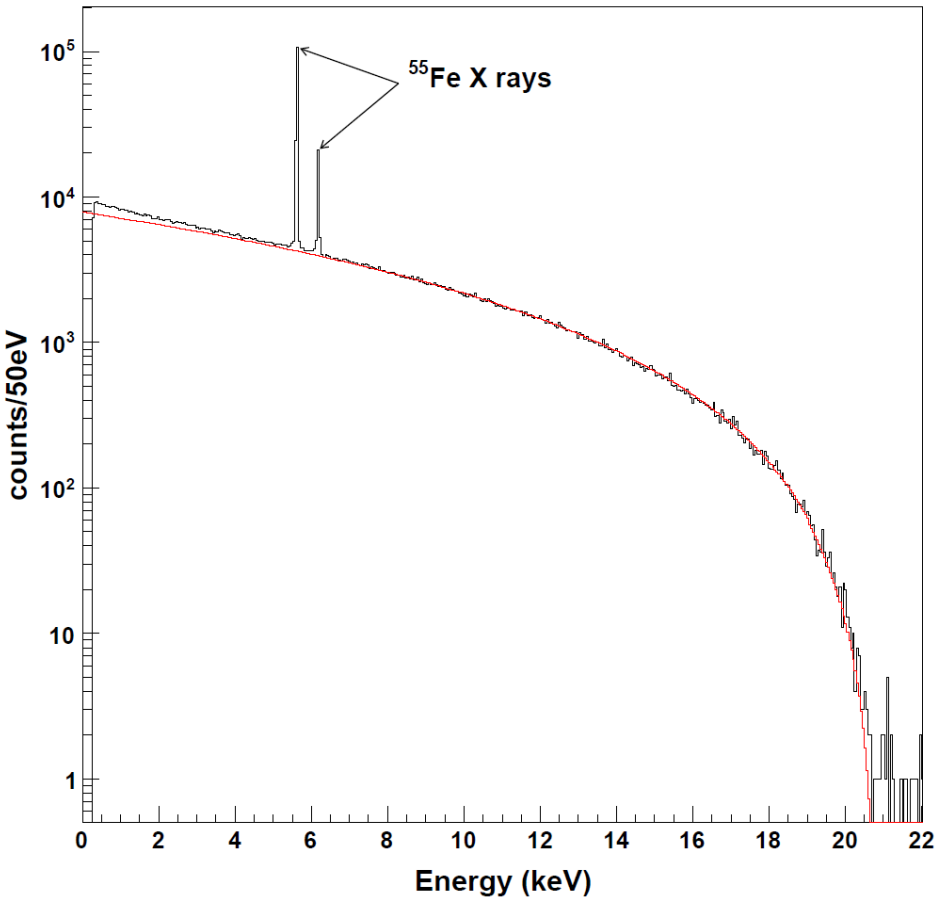

Figure 2 – Beta spectrum of ^{241}Pu measured with a metallic magnetic calorimeter and compared with a calculation from BetaShape. The forbidding order of the transition is first non unique, calculated as an allowed one, with an end-point energy of 20.8 keV. The theoretical spectrum was normalized by integration between 7 and 18 keV. An external source of ^{55}Fe was used for the energy calibration: the X-rays come from the Kα and the Kβ X-rays of the Mn daughter atom.

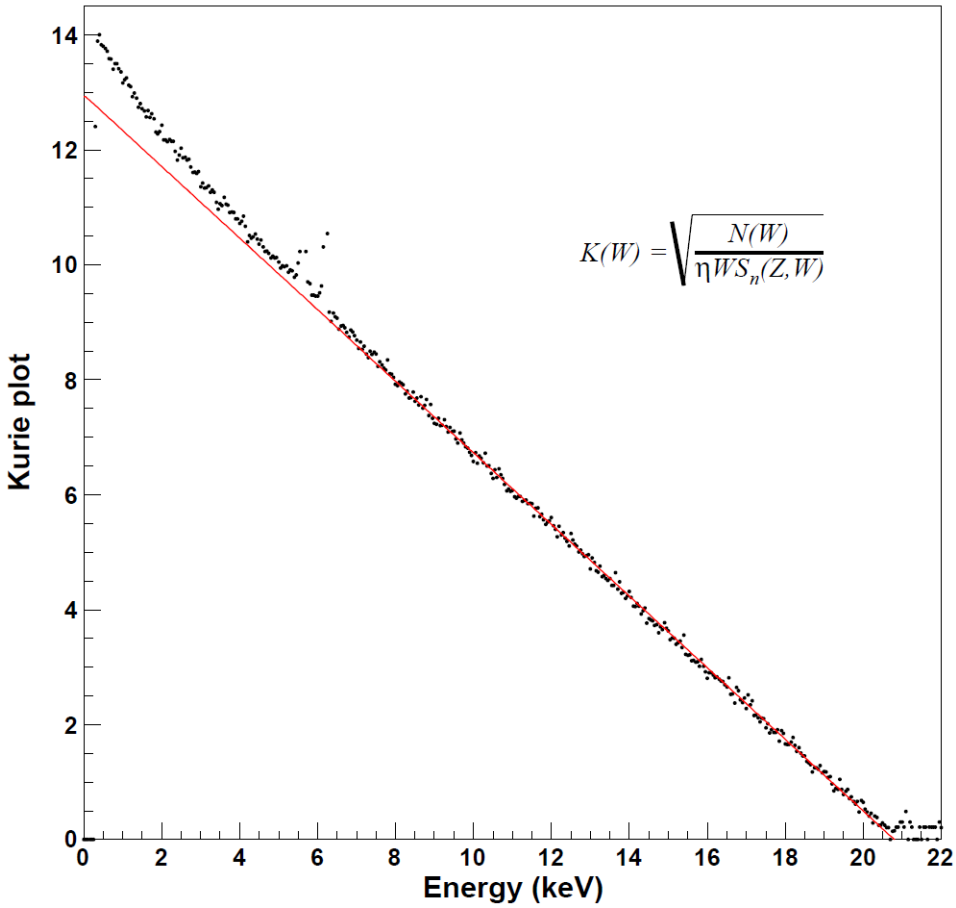

Figure 3 – The corresponding Kurie plot of Figure 2. The agreement between the measured spectrum and the BetaShape calculation is good over a wide range, except below 5 keV.

Tables

Allowed	(β^-) ^{3}H, ^{14}C, ^{24}Na, ^{32}P, ^{33}P, ^{35}S, ^{41}Ar, ^{45}Ca, ^{46}Sc, ^{60}Co, ^{63}Ni, ^{95}Nb, ^{106}Ru, ^{114}In, ^{133}Xe, ^{203}Hg (β^+) ^{11}C, ^{13}N, ^{15}O
First non unique forbidden	(β^-) ^{79}Se, ^{91}Y, ^{143}Pr, ^{147}Pm, ^{151}Sm, ^{176}Lu, ^{206}Tl, ^{210}Bi, ^{241}Pu
First unique forbidden	(β^-) ^{85}Kr, ^{89}Sr, ^{90}Sr, ^{90}Y
Second non unique forbidden	(β^-) ^{36}Cl, ^{99}Tc, ^{129}I
Second unique forbidden	(β^+) ^{26}Al
Third unique forbidden	(β^-) ^{40}K
Fourth non unique forbidden	(β^-) ^{113}Cd

Table 1 – List of the 38 transitions calculated for a comparison with the Radlist program. The non-unique transitions are calculated as unique transitions having the same variation of the total angular momentum.

Nucleus	Z	Transition order	Finite nuclear size correction	Screening correction	Radiative corrections greater than 1 % if
^{129}I	53	2nd non-unique	~ 0.44 %	~ 17 %	$E \geq 78\ \%\ E_{max}$
^{133}Xe	54	allowed	~ 0.007 %	~ 3 %	$E \geq 75\ \%\ E_{max}$
^{176}Lu	71	1st non-unique	~ 0.22 %	~ 4 %	$E \geq 72\ \%\ E_{max}$
^{203}Hg	80	allowed	~ 0.16 %	~ 4.5 %	$E \geq 58\ \%\ E_{max}$
^{241}Pu	94	1st non-unique	~ 0.03 %	~ 4.7 %	$E \geq 53\ \%\ E_{max}$

Table 2 – Magnitude of each correction applied in BetaShape for some nuclei. The non-unique transitions are calculated as unique transitions having the same variation of the total angular momentum.