

$^{137\text{m}}\text{Ba}$ - Comments on evaluation of decay data

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An evaluation of ^{137}Ba metastable state decay data has been performed by Sylvain Leblond during 2021-2023, with a literature cutoff date of January 2023.

Evaluation procedure and convention

The work presented follows mainly the DDEP evaluation procedures, with the associated uncertainty of any recommended value given at one standard deviation. If the authors of any of the references mentioned in this work use a different convention, the uncertainty of the reported measurement is adjusted to one standard deviation for consistency. Additionally, if any reported uncertainty is asymmetric, the value and uncertainty are re-defined symmetrically by means of the method described in [2012Au07].

1 Decay Scheme

^{137}Ba metastable state decays by internal gamma transitions to the ground state and to the first excited level of ^{137}Ba . Level spins and parities are taken from [2007Br23]. Level energies of the two excited states were evaluated in ^{137}Cs decay from measurements of the gamma transition energies to the ^{137}Ba ground state. These results are presented in Table 1, and the detailed analysis for each state can be found in the comment file of the DDEP ^{137}Cs decay recommendation [2023LeU0].

Table 1: Properties of ^{137}Ba states

Level	Spin-parity	Level energy (keV)
Ground state	$3/2^+$	0
1 st excited state	$1/2^+$	283.46 (7)
2 nd excited state	$11/2^-$	661.657 (3)

2 Nuclear Data

2.1 Half-life

The metastable ($11/2^-$) state of ^{137}Ba is mainly populated during the decay of ^{137}Cs and the resulting half-life has been extensively measured as a consequence of the ready availability of ^{137}Cs . An appropriate list of references is given in Table 2. Three studies have been rejected because of an absence of quantified uncertainties ([1959WaU0], [1963Ka34] and [2014KiU0]), while [1948To03] has been discarded due to lack of information regarding the uncertainty budget. Furthermore, [1966Mi10] is superseded by [1967Mi11].

Table 2: Half-life measurements of ^{137}Ba metastable state

Reference	Half-life (min)	Uncertainty (min)	Comments
1948To03	2.63	0.08	Discarded due to an absence of detail concerning the uncertainty
1959WaU0	2.50	-	Discarded: no uncertainty
1963Ka34	2.6	-	Discarded: no uncertainty
1965Me03	2.554	0.003	
1966Ma28	2.5513	0.0007	
1966Mi10	2.557	0.005	Superseded by 1966Mi11
1967Mi11	2.558	0.002 [#]	
1973LeZJ	2.5545	0.0007 [#]	
2014KwU0	2.6	-	Discarded: no uncertainty

[#] Uncertainty not given at 65% CL - adjusted appropriately in this work.

The resulting dataset is inconsistent ($\chi^2 = 5.54 / \chi^2_{\text{crit}} = 3.79$), and the Lweight algorithms generate a weighted average for which the uncertainty has been extended to cover the value assigned to the studies of [1966Ma28]: $T_{1/2} = 2.5532$ (19) min. Approximately 90% of the weighting is identified with [1966Ma28] and [1973LeZJ], although both references contain uncertainties that appear to be significantly underestimated (as both measurements are strongly inconsistent with each other). Therefore, my recommendation would be to adopt the unweighted average of the four measurements associated with the external uncertainty: **$T_{1/2} = 2.5545$ (13) min**, which is in good agreement with former evaluations for DDEP and ENSDF of $T_{1/2} = 2.552$ (1) min ([2006HeZT] and [2007Br23]).

2.2 Gamma Transitions

A summary of the recommendations for the gamma transitions observed in the decay of metastable ¹³⁷Ba is given in Table 3. The energies of these gamma transitions and associated uncertainties have been calculated from the properties of the level scheme of ¹³⁷Ba, as described in Section 1.

Table 3: Summary of recommended gamma transition properties

Initial state	Final state	Transition energy (keV)	Emission intensity (%)	Multipolarity
11/2 ⁻	3/2 ⁺	661.657 (3)	89.90 (13)	M4
11/2 ⁻	1/2 ⁺	378.20 (7)	1.06 (9) x 10 ⁻⁵	E5
1/2 ⁺	3/2 ⁺	283.46 (7)	1.91 (10) x 10 ⁻⁵	M1+E2

2.2.1 Intensity

Experimentally, the ^{137m}Ba decay has mainly been observed through the beta decay of ¹³⁷Cs. From the measurements, the decay is strongly dominated by the gamma transition from the 11/2⁻ metastable state to the ground state of ¹³⁷Ba. Alternatively, the metastable state can decay through gamma transition to the 1/2⁺ excited state, which subsequently decays to the ground state of ¹³⁷Ba. The two measured gamma emission intensities corresponding to this alternative decay path are much smaller than the gamma emission intensity of the main path. As a consequence, I recommend a 100% gamma transition intensity from the metastable state to the ground state, while the others two transition intensities are based on the balance of the intensities going in and out of the 1/2⁺ state. This choice ensures a complete decay scheme, including both decay paths from the metastable state of ¹³⁷Ba, while keeping the experimental measurements. The intensities deduced with such methods are listed in Table 4.

Table 4: Comparisons between recommended gamma transition and gamma-ray emission intensities

Initial state	Final State	Transition intensity (%)	Emission intensity (%)
11/2 ⁻	3/2 ⁺	100.0 (2)	89.90 (13)
11/2 ⁻	1/2 ⁺	2.02 (20) x 10 ⁻⁵	1.06 (9) x 10 ⁻⁵
1/2 ⁺	3/2 ⁺	2.02 (20) x 10 ⁻⁵	1.91 (20) x 10 ⁻⁵

It should be noted that the sum of the gamma transition intensities leaving the $11/2^-$ metastable is therefore $100.000\,020\,2 \pm 0.2\%$. This value is meaningful if taking the uncertainty into account but numerically higher than 100%. For users requiring 100% intensity out of the metastable state, I would recommend to neglect the alternative and very weak decay path and to use only the direct gamma transition to the ground state with 100% intensity.

2.2.2 Conversion Coefficients

Internal conversion coefficients have been calculated by means of the BrIcc version 2.2 code developed by Kibédi, *et al.* [2008Ki07]. Experimental ICC data are only available for the 661.657-keV transition, and an analysis was undertaken to validate the relevant BrIcc calculations. Details of the assessment of the experimental data are available in the comment file of the DDEP ^{137}Cs decay recommendation [2023LeU0].

The multipolarity of each gamma transition is determined from the spins and parities assigned to the initial and final states depopulated and populated, respectively. Such a procedure can be confirmed for the 661.657-keV gamma transition from experimental data that validate the conversion coefficients chosen. As detailed in the DDEP ^{137}Cs decay recommendation [2023LeU0], the BrIcc calculations performed assuming a pure M4 transition type fit very well with the recommendations obtained from the experimental data.

3 Atomic Data

Fluorescence yields were calculated by means of the SAISINUC program ([2008DuZX]) which incorporates the data of Schönfeld ([1996Sc06]). X-ray and Auger-electron energies were also determined within the SAISINUC program, based on input data from [1999ScZX] and [1998ScZM], respectively. The 2013 version of the EMISSION code was used to calculate the X-ray emission probabilities, as described in [2000Sc47] and implemented by SAISINUC in terms of the evaluated gamma ray emission probabilities and conversion coefficients.

4 Emissions

4.1 Photons

The energies of the gamma-ray emissions were calculated from the energies of the gamma transitions, taking into account the gamma recoil energy of ^{137}Ba (see Section 1), and are given in Table 5.

Table 5: Energy comparison between gamma transitions and gamma-ray emissions

Initial state	Final state	Transition energy (keV)	Emission energy (keV)
$11/2^-$	$3/2^+$	661.657 (3)	661.655 (3)
$11/2^-$	$1/2^+$	378.20 (7)	378.20 (7)
$1/2^+$	$3/2^+$	283.46 (7)	283.46 (7)

Gamma-ray emission intensities have been determined from analyses of the measurements for each transition, and are detailed in the following subsections.

4.1.1 Gamma-ray emission at 661.657 keV: $11/2^- \rightarrow 3/2^+$

The main gamma emission has been measured mainly following the decay of ¹³⁷Cs. In this work, the intensity of the gamma emission has been calculated to ensure 100% transition intensity. The uncertainty on the intensity is similarly calculated based on the DDEP ¹³⁷Cs decay recommendation [2023LeU0].

4.1.2 Gamma-ray emission at 378.2 keV: $11/2^- \rightarrow 1/2^+$

The emission probability of this E5 gamma transition from the $11/2^-$ state to the $1/2^+$ state of ¹³⁷Ba was assumed to be very low. Nevertheless, experimental studies by Moran, *et al.* have succeeded in determining this gamma transition to be near-pure E5 with intensity $I_\gamma = 1.06 (9) \times 10^{-5}\%$ [2014Mo32]. Although the uncertainty budget lacks detail and is judged by the evaluator to be underestimated, this low-intensity gamma transition has been recommended in the proposed decay scheme of ^{137m}Ba.

4.1.3 Gamma-ray emission at 283.46 keV: $1/2^+ \rightarrow 3/2^+$

All the intensity measurements of the 283.46 keV gamma emission are made following the beta decay of ¹³⁷Cs. Thus, they include the beta feeding of $1/2^-$ and should not be used for the evaluation of the ^{137m}Ba decay scheme. As a consequence, the 283.46 keV gamma emission is deduced from the balance of the intensities from the $11/2^-$ state deexcitation.

4.1.4 Double-gamma emission from $11/2^-$ state of ¹³⁷Ba

¹³⁷Ba is one of the few nuclides for which double-gamma emission is sufficiently competitive to be measured. Effectively, the 661.657-keV $11/2^-$ state of ¹³⁷Ba can decay to the ground state via the simultaneous emission of two photon emissions, as well as by the much more probable single 661.657-keV gamma-ray. Two gamma-rays can be emitted simultaneously, sharing the energy corresponding to the transition between the two states. Hence, such a process do not produce a discrete energy spectrum but instead a continuous spectrum with a shape that depends on the phase space of the decay.

The first investigations of possible ¹³⁷Cs double-decay were performed in the 1960s and 1970s with the detection of two K-electrons [1960Be20, 1969Lj01, 1971Lj02]. While the first sole gamma-electron detection was performed at approximately the same time [1971Lj01], the first tentative detection of the two-gamma emission did not occur until the 1990s [1992Ba45]. Quite recently, the topic has drawn more interest and a series of measurements has been performed to detect the double-gamma emission [2015MeZZ, 2015Wa29, 2016PiZW]. The probability of such an event is extremely small, more than six orders of magnitude less probable than the direct single-photon emission [2015Wa29, 2016PiZW]:

$$I_{\gamma\gamma}(661.66 \text{ keV}) = 2.05 (37) \times 10^{-6} I_\gamma(661.66 \text{ keV}).$$

Given the nature of the double-gamma emission and the extremely low probability, this particular decay mode has not been included in the tabulated recommendations.

5 Prospects for improvements

The recommendation on the half-life of the ^{137}Ba is based only on four measurements, two of which seem to have underestimated uncertainty. I would therefore recommend new measurements of the metastable half-life.

The emission probabilities of two of the three gamma rays are extremely small, and only a limited number of references are relevant to the evaluation of the decay characteristics of the third gamma ray. The deduced decay scheme is consistent, given the uncertainties, but would largely benefit from additional measurements of the two gamma emissions of the ^{137}Ba metastable state.

6 References

All the publications used in this work are sorted below in terms of their associated NSR keynumbers [2011Pr03]. If no keynumber has been assigned to a paper, an NSR-type substitute has been attributed to the reference with U0 as the terminator. A full reference is given for each publication, and the type of information extracted is specified within square brackets.

1948To03	J. Townsend, <i>et al.</i> , Physical Review 74 (1948) 499 [$^{137\text{m}}\text{Ba}$ half-life]
1959WaU0	R.H. Wasserman, <i>et al.</i> , Science 129 (1959) 568-569 [$^{137\text{m}}\text{Ba}$ half-life]
1960Be20	W. Beusch, Helvetica Physica Acta 33 (1960) 363-392 [Double gamma decay]
1963Ka34	T. Kaminishi, C. Kojima, Japanese Journal of Applied Physics 2 (1963) 399-405 [$^{137\text{m}}\text{Ba}$ half-life]
1965Me03	J.S. Merritt, J.G.V. Taylor, Analytical Chemistry 37 (1965) 351-354 [Beta-decay intensity, α_{T} coefficient, α_{K} coefficient, Gamma-ray intensity, ^{137}Ba half-life]
1966Ma28	P.G. Marais, <i>et al.</i> , Journal of the South African Chemical Institute 19 (1966) 1-5 [$^{137\text{m}}\text{Ba}$ half-life]
1966Mi10	V. Middelboe, Nature 211 (1966) 283-284 [$^{137\text{m}}\text{Ba}$ half-life]
1967Mi11	V. Middelboe, Matematisk-fysiske Meddelelser det Kongelige Danske Videnskabernes Selskab 35 (1966) 1-19 [$^{137\text{m}}\text{Ba}$ half-life]
1969Lj01	A. Ljubicic, <i>et al.</i> , Physical Review 187 (1969) 1512 [Double gamma decay]
1971Lj01	A. Ljubicic, <i>et al.</i> , Physical Review C 3 (1971) 824 [Double gamma decay]

1971Lj02	A. Ljubicic, <i>et al.</i> , Physical Review C 3 (1971) 831 [Double gamma decay]
1973LeZJ	J. Legrand, <i>et al.</i> , CEA-R-4428 (1973) [Gamma intensity, Beta decay intensity, α_T coefficient, α_K coefficient, K/LM coefficient ratio, ^{137m} Ba half-life]
1992Ba45	V.K. Basenko, <i>et al.</i> , Bulletin of the Russian Academy of Sciences: Physics 56 (1992) 94 [Double gamma decay]
1996Sc06	E. Schönfeld, H. Janssen, Nuclear Instruments and Methods in Physics Research A 369 (1996) 527 [Atomic data]
1998ScZM	E. Schönfeld, G. Rodloff, PTB-6.11-98-1 (1998) [¹³⁷ Ba Auger electron energies]
1999ScZX	E. Schönfeld, G. Rodloff, PTB-6.11-1999-1 (1999) [¹³⁷ Ba X-ray emission probabilities]
2000Sc47	E. Schönfeld, H. Janssen, Applied Radiation and Isotopes 52 (2000) 595 [¹³⁷ Ba X-ray and Auger emission probabilities]
2006HeZT	R.G. Helmer, V.P. Chechev, LNE-LNHB CEA/Table of Radionuclides (2006) [DDEP evaluation]
2007Br23	E. Browne, J.K. Tuli, Nuclear Data Sheets 108 (2007) 2173-2318 [ENSDF evaluation]
2008DuZX	C. Dulieu, <i>et al.</i> , Proceedings of the International Conference on Nuclear Data for Science and Technology, 22-27 April 2007, Nice, France (2008) 97-99 [SAISINUC]
2008Ki07	T. Kibédi, <i>et al.</i> , Nuclear Instruments and Methods in Physics Research A 589 (2008), 202 [Conversion coefficients]
2011Pr03	B. Pritychenko, <i>et al.</i> , Nuclear Instruments and Methods in Physics Research A 640 (2011) 213 [NSR]
2012Au07	G. Audi, <i>et al.</i> , Chinese Physics C 36 (2012) 1157-1286 [Asymmetric uncertainty calculation]
2014KwU0	D.C. Kweon, <i>et al.</i> , Journal of the Korean Physical Society 65 (2014) 532-540 [¹³⁷ Ba half-life]
2014Mo32	K. Moran, <i>et al.</i> , Physical Review C 90 (2014) 041303 [Gamma-ray intensity]

- 2015MeZZ** E. Merchan, *et al.*, CGS15 Capture Gamma-Ray Spectroscopy and Related Topics, Dresden Germany, 25-29 August 2014, EPJ Web of Conferences 93 (2015)
[Double gamma decay]
- 2015Wa29** C. Walz, *et al.*, Nature 526 (2015) 406
[Double gamma decay]
- 2016PiZW** N. Pietralla, *et al.*, XXI Int. School Nucl. Phys. Applications, and Int. Symp. on Exotic Nuclei (ISEN-2015), 6-12 September 2015, Varna, Bulgaria, Journal of Physics: Conference Series 724 (2016) 012039
[Double gamma decay]
- 2023LeU0** S. Leblond, LNE-LNHB/CEA Table of Radionuclides: DDEP evaluation of ^{137}Cs decay data, July 2023
[Evaluation of ^{137}Cs]