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On the decay of the $J^{\pi} = 11/2^{-}$, $T_{1/2} = 38.9$ h, isomer in ¹³³Ba: Search for the E5 transition and verification of the EC branch

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Abstract. This paper presents a search for the competing $11/2^- \rightarrow 1/2^+$ E5 branch to the main $11/2^- \rightarrow 3/2^+$ M4 transition from the $T_{1/2} = 39$ h, $J^{\pi} = 11/2^-$ isomeric state in ¹³³Ba. An upper limit of $1.8 \cdot 10^{-5}$ could be established for the E5/M4 intensity ratio. In addition a long-standing controversy in the EC-decay of the same isomer was resolved: only one EC branch to the $11/2^+$ state in ¹³³Cs was observed. The shell-model structures of the involved states in ¹³³Ba and ¹³³Cs were studied with the microscopic quasiparticle-phonon model (MQPM). Good agreement with the decay rates $B(\lambda)$ was obtained. In particular, the B(M4) value is well reproduced and further only one low-lying 11/2 state could be identified in the latter nuclide, supporting the experimental results.

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1. Introduction

The isomeric $11/2^{-}$ level in ¹³³Ba has been known for several decades. It was first observed as a (40.0 ± 0.5) h isomer by Cork and Smith 1941 [1], although in that study the spin was wrongly assigned to 9/2. After this it has been studied by several others, see references [2, 3, 4, 5, 6]. The half-life of the isomeric state is $T_{1/2} = 38.9$ h and the main M4 decay of the level proceeds to an excited $3/2^{+}$ state at 12 keV [7]. A weak branch to the $1/2^{+}$ ground state, of multipolarity E5, was reported in an Annual Report of Tokyo University [6]. Furthermore, the $11/2^{-}$ isomer has a weak EC decay branch to 133 Cs, $I_{EC} = 0.0096(11)$, notably to an $11/2^{+}$ level at 633 keV. An earlier work claims



Figure 1. Level scheme of ¹³³Ba illustrating the M4/EC decay of the $T_{1/2}$ =38.9 hours $J^{\pi} = 11/2^{-}$ isomer, and the possible E5 decay branch. The isomeric EC decay branch to the $J^{\pi} = 11/2^{+}$ state in ¹³³Cs has an intensity of 0.011%. The other branch, to the $J^{\pi} = 11/2^{-}$ state, is under dispute. The 10.7 yr EC decay of the ¹³³Ba ground state is included for completeness (its accompanying γ -rays will be discussed in the text). The level scheme is based on [7, 8].

decay to both this level and another almost degenerate $11/2^{-}$ level at 605 keV, cf. [5, 8] but this latter claim is not supported in [6]. These two decay paths (γ -ray and EC) are marked with question marks in figure 1 and discussed in more detail in section 2.

This paper presents an investigation on the competing E5 branch $11/2^- \rightarrow 1/2^+$ to the main $11/2^- \rightarrow 3/2^+$ M4 in ¹³³Ba. Furthermore, the study aims at settling the issue of the existence of the $11/2^-$ level in ¹³³Cs, suggested in reference [8]. This level should also be fed by the EC decay of the 38.9 h isomer in ¹³³Ba. The isomeric state in ¹³³Ba was populated using the reaction ¹³³Cs(p,n)¹³³Ba and a CsI target.

In order to understand the transition rates related to the microscopic structures of the involved states, we have performed calculations within a QRPA (quasiparticle random-phase approximation)-based approach of the microscopic quasiparticle-phonon model (MQPM) [9, 10]. These theoretical studies are used to shed light on the results and support the experimental findings on the two issues that have raised controversy:

1) Despite the high multipoles of the transitions $11/2^- \rightarrow 3/2^+$ and $11/2^- \rightarrow 1/2^+$, the $11/2^-$ state decays predominantly by γ -ray emission [11]. The latter seems to be much more hindered than the former. We intend to study the existence of the E5 transition.

2) Does the $11/2^{-}$ level exist in the level structure of ¹³³Cs, and what are the EC decay strengths to the $11/2^{-}$ and/or $11/2^{+}$ levels?

2. Experimental method

The Tandem van de Graaff at the Accelerator Laboratory at Helsinki University was used to determine a suitable particle energy for the reaction ${}^{133}\text{Cs}(p,n){}^{133}\text{Ba}$ to populate the $11/2^-$ state in ${}^{133}\text{Ba}$. The advantage of using the Tandem van de Graaff is that the energy can be changed rapidly but the disadvantage is that it does not reach sufficiently high in energy. In the experiment a proton beam with the average current of 50 nA and five different energies; 5.0 MeV, 6.2 MeV, 7.4 MeV, 8.6 MeV and 9.8 MeV was used. The proton irradiation of a thin CsI crystal target resulted in two different reactions; the desired reaction producing ${}^{133}\text{Ba}$ and the reaction ${}^{127}\text{I}(p,n){}^{127}\text{Xe}$. Singles γ -rays were measured online with a Ge-detector perpendicular to the direction of the particle beam.

To avoid the relatively long decay time for the isomer $11/2^{-1}$ in ¹³³Ba the production rate for the $13/2^{-}$ state at 902 keV in Ba was studied. This state decays solely with 613.6 keV gamma emission to the $11/2^{-}$ state and thus it gives a minimum of the production rate of the $11/2^{-}$ state. The experimental production rate was normalised by setting the production rate to 1 at the incident energy of 5.0 MeV, see figure 2. From the experimental values the excitation function for the $11/2^{-}$ state was estimated to have a maximum at an incident energy of about 12 MeV. This was confirmed by an excitation function calculated with the software package TALYS [12] giving a maximum at 10.5 MeV and by the results in a recent study by F. Tárkányi et al. [13], as well as in two older ones [14, 15]. The fact that the accompanying reaction ${}^{127}I(p,n){}^{127}Xe$ shows a cross section maximum at the same energies [16] is not a problem as the halflife of the $9/2^{-}$ isomeric state in ¹²⁷Xe is 69.2 s, which is short compared to that of the the isomeric state in ¹³³Ba. Furthermore, the decay of the ¹²⁷Xe ground state through electron capture, $T_{1/2} = 36.4$ d, does not have disturbing secondary γ -rays [17]. Figure 2 shows experimental data from this work and from references [13, 15] as well as the calculated data.

The actual activation study of the $11/2^{-}$ state in ¹³³Ba was done at the Accelerator Laboratory at Åbo Akademi. The target, a $(10 \times 10 \times 2)$ mm³ CsI crystal, was placed in a water cooled target holder with a 25 μ m stainless steel foil in front of it. The foil degrades the particle energy with 0.49 MeV. The target was irradiated with a 1 μ A, 12 MeV proton beam for 3 hours. After the irradiation the target was removed from the holder and the dose rate of the target was measured to 4 mSv/h at a distance of 5 cm.

The decay of the activated CsI target was measured during a period of 11 days. Singles gamma-ray were recorded using one high-purity Ortec Ge detector of 19% efficiency. During the first 84 hours the source was placed 1 m from the detector to keep to a count rate between 1000 cps at the beginning of the measurement and 600 cps at the end. The low count rate reduces the dead time in the measurement system. Assuming a nonparalyzable model [18] for the dead time, the ratio between the dead-time-corrected count rate and the measured count rate was 1.0101 for the highest measured count rate of 1000 cps and a pulse width of 10 μ s. On the average the ratio between the length of



Figure 2. The figure shows a plotted experimental data from this work (\triangle) and the cross sections from F. Tárkányi et al. [13] (×), Sakamoto et al. [15] (□), as well as the calculated data (- - - -)

the measurement interval and the live time of the system was 1.0099 and thus the live time is well corrected for the dead time. After the half-life measurement the target was moved closer to the detector and the measurement was continued with a count rate of 7000 cps to get higher statistics. A spectrum collected during the first 12 hours after the activation is showed in figure 3. Strong peaks from the decay of the isomeric state in ¹³³Ba, 276 keV, and the decay of the ground state of ¹²⁷Xe can be seen in the upper panel. The peak origin from the decay of the $11/2^+$ state at 633 keV to the ground state in ¹³³Cs can be found in the lower panel.

3. The decay of the $11/2^{-}$ isomer

The half-lives of the $11/2^{-}$ state in ¹³³Ba have been reported by several authors, see table 1. In this work the half-life was determined from the M4 γ -decay. The activity measured during time intervals of 12 h for the first 72 hours were used to determine the half-life. The total counts where divided with the live time of the measuring system and corrected to the beginning of the time interval. The half-life and the statistical error, $T_{1/2} = (38.92 \pm 0.09)$ h, was obtained from an exponential fit to the values using gnuplot 4.2.0, see figure 4. The systematical error is expected to be of the same order of magnitude or lower than the statistical one. The result clearly confirms the previous results by Wille et al. but the shorter half-life determined by Ando et al. drops outside the error limits.

Sugiyama and Kikuchi [8] have reported electron-capture decays to two levels at 633 keV and 605 keV in 133 Cs in addition to the 275.6 keV M4 transition in 133 Ba. These two EC-branches where identified by detection of gamma-rays corresponding to



Figure 3. A spectrum collected during the first 12 hours after the activation. The strongest peak origins from the decay of the isomeric state in 133 Ba. Strong peaks originating from the decay of the ground state of 127 Xe can also be found.

Table 1. Reported half-lives for the isomeric $11/2^-$ state ¹³³Ba and intensity ratios E5/M4.

$T_{1/2}\left[h\right]$	Intensity ratio $E5/M4$	References
$\begin{array}{l} 38.84 \pm 0.11^a \\ 40.0 \pm 0.5 \\ 42.5 \\ 38.9 \pm 0.1 \\ 38.05 \pm 0.04 \end{array}$	$< 1.8 \cdot 10^{-5}$ $(3.6 \pm 2.5) \cdot 10^{-4}$	This work Cork [1] Hill [2] Wille [3] Ando [6]

 $^{^{}a}$ Purely statistical error, the systematic error is expected to be of the same order of magnitude or lower than the statistical one.

the two transitions to the ground state. The relative intensities for both transitions were given as 0.011 % and 0.013 % and the log ft values as 7.9 and 8.0, respectively.



Figure 4. The figure shows the decrease of the corrected activity corresponding to the decay $11/2^{-}$ state in ¹³³Ba as a function of time. The line is fitted to the data.

In another work by Kikuchi [5] both levels in ¹³³Cs have been assigned spin and parity. The assignments were based on angular distribution measurement of the radiation from the reaction ¹³³Cs(n, n' γ). The level at 633 keV was assigned the spin and parity $11/2^+$ and was considered to be based on the $g_{9/2}$ proton state coupled to the lowest 2^+ phonon state in ¹³²Xe. The other level at 605 keV was assigned spin $11/2^-$ from the $h_{11/2}$ proton state.



Figure 5. A part of the spectrum showing that it is possible to fit a peak at 605 keV. The spectra were measured during 84 h. The inserted figure shows the growth of the peak at 605 keV (\times) and 633 keV (*) as a function of time. The line is fitted to the data.

In order to study the 605 keV line the position of the fitted peak was locked to the correct position and the shape was obtained from near-by peaks. But the peak did not

grow with increasing recording time in the same manner as the peak for the 633 keV transition which is known to be fed from the EC-decay of 133m Ba, see figure 5 and the insert in the figure. The peak at 605 keV grows, without significant deviations, linearly in time which indicates that it originates from a long-lived nucleus in the background. However, the source could not be identified. Furthermore, the $11/2^- \rightarrow 11/2^-$ transition should be faster since it corresponds to an allowed EC decay whereas the $11/2^- \rightarrow 11/2^+$ transition corresponds to a first-forbidden EC decay. Thus, if the unidentified peak at 605 keV would origin from the decay of a $11/2^-$ state in 133 Cs it should be larger than the peak at 633 keV. However, in this study the intensity ratio of the former to the latter was found to be smaller than 0.12.

Ando et al. report a weak E5 transition between the isomeric $11/2^{-}$ state and the $1/2^+$ ground state in ¹³³Ba. The relative intensity was found to be $(3.6 \pm 2.5) \cdot 10^{-4}$ of the 276 keV M4 transition [6]. In the present study spectra collected during the first 72 h after the activation were used in the analysis. With the relative intensity given above the peak area at 288 keV should be about $9.7 \cdot 10^3$ counts as there were $2.70 \cdot 10^7$ counts in the 276 keV peak. However, a clear peak cannot be found in the spectrum. To establish an upper limit for the intensity a peak, with the shape and background parameters locked, was forced at the position of the assumed 288 keV peak. Figure 6 shows the forced-peak fit at 288 keV and fits to the near-lying peaks at 295 keV and 303 keV. The same background and parameters for peak shape were used for all three peaks. The forced peak height 125 counts and the width 3.0 channels resulted in a peak area of 490 counts and a peak at this position would be smaller. As the internal conversion coefficients for both the M4 and the E5 transitions are of the same order [19], 4.64 and 4.08 respectively, internal conversion can be neglected. This gives an upper limit of $1.8 \cdot 10^{-5}$ for the E5/M4 intensity ratio. The ratio between the calculated transition probabilities is $3.1 \cdot 10^{-6}$, see table 2, in agreement with the experimental upper limit.

4. Theoretical calculations

In this paper the MQPM [10] was used for theoretical calculations of the wave functions and excitation energies of ¹³³Ba and ¹³³Cs. In the model the three-quasiparticle states of an odd-A nucleus are formed by coupling one-quasiparticle states to QRPA (quasiparticle random-phase approximation [20]) phonons. The MQPM calculations were performed in four steps:

First the single-particle energies were calculated from the Coulomb-corrected Woods-Saxon potential with the Bohr-Mottelson parametrization [21]. In the calculations a valence space containing 11 neutron orbitals from $0f_{7/2}$ to $0h_{11/2}$ and 9 proton orbitals from $1p_{1/2}$ and $0h_{11/2}$ was used. The calculated results could be improved by slightly adjusting the proton and neutron single-particle energies in the vicinity of the respective Fermi surfaces.

BCS calculations were then performed separately for neutrons and protons. The Bonn one-boson-exchange potential [22] was used as the two-body interaction. The



Figure 6. A fit to the peaks close to the position of the 288 keV peak. The assumed peak at 288 keV was created by force with the peak and background shape fixed based on the analysis of the peak at 275 keV. The forced peak height was 125 counts and the width 3.0 channels

experimental neutron and proton pairing gaps were calculated from the three-point formulae (see e.g. [23]). The monopole matrix elements were renormalized by adjusting the proton and neutron pairing strengths [24] so that the lowest one-quasiparticle energy was approximately equal to the experimental pairing gap.

The QRPA model was subsequently used to calculate the excitation spectrum of the even-even reference nucleus (¹³⁴Ba). For each multipole J^{π} the particle-particle ($g_{\rm pp}$) and the particle-hole ($g_{\rm ph}$) strengths were adjusted so that the experimental low-lying energy spectra were reproduced.

In the final step of the MQPM calculations the interaction Hamiltonian was diagonalized (see [10]) to obtain the MQPM wave functions and excitation energies. A phonon basis containing all QRPA states having angular momentum $J \leq 10$ ($\pi = +, -$) and excitation energy $E_{\omega} \leq 3.5$ MeV was used. The obtained energy spectra are compared with experimental data in figures 7 and 8 and the main configuration of selected states are given in table 2. As seen in the figures, the agreement is very good and the calculations does not support two 11/2 states at about 600 keV in ¹³³Cs.

Furthermore, electromagnetic transition probabilities and half-lives have been calculated for the γ -transitions shown in figure 1. The calculated partial half-life for the M4 transition, using free-nucleon gyromagnetic ratios, is $0.5027 \cdot 10^5$ s \approx 14.0 h. However, effective gyromagnetic ratios of $\tilde{g}_l^{\nu} = -0.07$, $\tilde{g}_s^{\nu} = -2.30$ and $\tilde{g}_l^{\pi} = 1.13$, $\tilde{g}_s^{\pi} = 3.546$ from [25] for the neutrons and protons, respectively, gave $T_{1/2} = 0.1426 \cdot 10^6$ s ≈ 39.6 h, in excellent agreement with the experimental value. The calculated partial half-life of a possible E5 admixture $(1.64 \cdot 10^{11} \text{ s})$ supports the findings in reference [4] that this transition is of pure M4 character.

For the suggested 288 keV E5 transition from the $11/2^{-}$ state to the ground state



Figure 7. Theoretical and experimental states of 133 Ba. The experimental data is obtained from [7].



Figure 8. Theoretical and experimental states of 133 Cs. The experimental data is obtained from [7].

the partial half-life was calculated using effective charges $e_{eff}^{\nu} = \chi e$ and $e_{eff}^{\pi} = (1 + \chi)e$ with $\chi = Z/A$ [23] i.e. 0.421e and 1.421e for the neutron and proton respectively, resulting in $T_{1/2} = 4.563 \cdot 10^{10}$ s = $1.27 \cdot 10^7$ h and branching of $3.1 \cdot 10^{-6}$ as mentioned above. The effective gyromagnetic ratios and effective charges may origin from meson exchange currents and core polarizations, respectively, see reference [25] for details.

The calculated partial half-lives for the other transitions in ¹³³Cs and ¹³³Ba are in good agreement with the experimental half-lives of the states with some exceptions. The low energy transition of 12 keV between the $3/2^+$ and $1/2^+$ states in ¹³³Ba is an *l*-forbidden M1 transition with a low E2 admixture [4]. A similar argument explains the overestimated half-life for the 53 keV transition between the $1/2^+$ state and the $3/2^+$ state and the 81 keV transition between the low-lying $5/2^+$ state and the ground state, both *l*-forbidden M1 transitions [26]. In these cases the calculations gives the realistic partial half-lives for the M1 transitions; $9.25 \cdot 10^{21}$ s, $3.00 \cdot 10^{27}$ s and $3.46 \cdot 10^{21}$ s, respectively. This is because the three-quasiparticle amplitudes of the MQPM states in these cases are very small and thus leading to an almost vanishing M1 matrix element. The results are summarized in table 2.

comparison with experimental data from reference	, calculated using effective gyromagnetic ratios and	l in the calculations are shown.
Table 2. Summary of MQPM results for the interesting states in ¹³³ Cs and ¹³³ Ba and comparison with experimental data fr	[7]. The experimental $T_{1/2}$ values are half-lives of the states whereas theoretical values, calculated using effective gyromagnet	charges, are the partial half-lives of the transitions. Only the main configurations used in the calculations are shown.

		Main	Energy	[keV]			Ι	Jecay		
	State	Conf	theory	exp.	$T_{1/2}$ [s]	$E_{\gamma} \; [{\rm keV}]$	final state	exp. multipole	$T^{th}_{1/2} \; \mathrm{[s]}$	th. multipole
^{133}Cs	$\frac{7}{2^+}$	g7/2 dr./0	g.s. 16.5	8.8. 	$6.28 \cdot 10^{-9}$	2	o, J	M1+F2	$8.37\cdot10^{-5}$	E.2
	1	7/00		H D		ł	2//2	-	$3.46 \cdot 10^{21}$	M1
	$5/2^+$	$\mathrm{g}_{7/2}\otimes 2^+$	606	161	$1.72\cdot 10^{-10}$	80	$d_{5/2}$	M1+E2	$5.52\cdot 10^{-6}$	M1
							- (-		$7.51\cdot 10^{-5}$	E2
						161	$g_{7/2}$	M1+E2	$1.02\cdot 10^{-7}$	M1
									$1.99\cdot 10^{-7}$	E2
	$3/2^+$	$d_{3/2}$	577	384	$2.1\cdot 10^{-11}$	223	$\mathrm{g}_{7/2}\otimes 2^+$	M1+E2	$1.34\cdot 10^{-6}$	M1
							·		$3.89\cdot 10^{-6}$	E2
						303	$d_{5/2}$	M1+E2	$1.73\cdot 10^{-12}$	M1
							×		$9.97\cdot 10^{-9}$	E2
						384	$g_{7/2}$	E2	$1.40\cdot 10^{-9}$	E2
									$2.61\cdot 10^{21}$	M1
	$1/2^+$	$\mathrm{S}_{1/2}$	638	437	$\leq 1.50\cdot 10^{-10}$	53	$d_{3/2}$	M1+E2	$8.83\cdot 10^{-6}$	E2
							×		$3.00\cdot 10^{27}$	M1
						276	$\mathrm{g}_{7/2}\otimes 2^+$	E2	$2.48\cdot 10^{-6}$	E2
							I		$3.70\cdot 10^{-7}$	M1
						356	$d_{5/2}$	E2	$5.59\cdot10^{-10}$	E2
									$8.79\cdot 10^{21}$	M1
	$11/2^+$	$\mathrm{g}_{7/2}\otimes 2^+$	795	633	$5.4\cdot10^{-12}$	633	$g_{7/2}$	E2	$2.06\cdot 10^{-9}$	M1
									$6.73\cdot 10^{-10}$	E2
^{133}Ba	$1/2^+$	$\mathrm{S}_{1/2}$	o.s.	s. S						
	$3/2^{+}$	$d_{3/2}$	50	12	$7.0\cdot 10^{-9}$	12	$S_{1/2}$	M1	1.07	E2
		×							$9.25\cdot 10^{21}$	M1
	$11/2^-$	$\mathrm{h}_{11/2}$	302	288	$1.40\cdot 10^5$	276	$d_{3/2}$	M4	$1.43\cdot 10^5$	M4
									$1.64\cdot 10^{11}$	E5
						288	S1/9	E5	$4.56 \cdot 10^{10}$	E5

5. Conclusions

Both the EC- and the γ -decay branches of the isomeric $11/2^{-}$ state in ¹³³Ba have been studied. The proposed competing EC-decay branch to a $11/2^{-}$ level at 605 keV in ¹³³Cs was not found, even though it was possible to fit a small peak at the correct position in the obtained energy spectrum. However, the EC branch to the $11/2^{+}$ level at 633 keV was confirmed. The peak at 605 keV behaves as if it would originate from a long-lived source in the background. Furthermore, the MQPM calculation gives an $11/2^{+}$ state at 795 keV with the main configuration arising from a proton $g_{7/2} \otimes 2^{+}$ phonon and, in addition a $11/2^{-}$ state at 952 keV with the main configuration $h_{11/2}$. Corresponding levels are also found experimentally at 633 keV and 1071 keV, respectively [7]. Thus the existence of a $11/2^{-}$ state at 605 keV is not supported by experiment nor by MQPM calculations. Therefore, the only existing EC-decay from the isomeric state in ¹³³Ba goes to the level at 633 keV in ¹³³Cs.

The weak E5 γ -transition to the ground state in ¹³³Ba could not be detected. Therefore, only an upper limit for the intensity relative to the main γ decay branch at 276 keV could be established to $2.2 \cdot 10^{-5}$. The theoretical calculation suggests that the E5 transition should be a single-particle transition $h_{11/2} \rightarrow s_{1/2}$ and the M4 transition should have the single-particle structure $h_{11/2} \rightarrow d_{3/2}$. The suppression of the E5 transition relative to the M4 transition is thus simply the suppression arising from the higher multipolarity $\lambda = 5$ of the electric transition, as can be simply estimated by using the formalism described in [23].

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