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Reduction in the uncertainty of the neutron-capture cross section of ^{210}Bi: Impact of a precise multipolarity measurement of the $2^- \rightarrow 1^-$ main ground-state transition

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Abstract. The mixing ratio of the main 320-keV, $M1 + E2$ ground-state γ transition in ^{210}Bi has been more precisely quantified, allowing a significant reduction in the uncertainty of measurements of the neutron-capture cross section to the ground state of ^{210}Bi from 25% to 0.9%. Accurate values for neutron-capture cross sections to both the ground and long-lived 9^- isomeric state at 271 keV in ^{210}Bi are of particular importance as Pb-Bi finds increased usage in Accelerator Driven Systems.

1. Introduction

The ^{210}Bi nucleus is a one-proton one-neutron particle system with respect to the doubly-magic ^{208}Pb core, therefore the investigations of its structure may deliver information on the properties of nuclei around closed shells. Moreover, studies of the ^{209}Bi(n,γ)^{210}Bi reaction are very important because Pb-Bi can be used as a coolant in fast reactor systems or as a spallation neutron-production target in Accelerator Driven Systems. The measurements of the neutron-capture cross section are of particular interest because the ^{209}Bi(n,γ)^{210}Bi process contributes significantly to the short- and long-term radiotoxicity of the material used.

The ^{210}Bi nucleus is populated by the neutron-capture reaction in the 4605-keV state, which then emits γ-ray cascades feeding the 1^- ground state or the long-lived 9^- isomer. Beta decay of the ^{210}Bi ground state with the half-life of 5.013 d produces the ^{210}Po nucleus, which as an α emitter with $T_{1/2} = 138$ d is a source of short-term radiotoxicity. On the other hand, due to the large spin difference with respect to the 1^- ground state, the second excited state with a spin and parity of 9^- decays by α emission with $T_{1/2} = 3.04 \times 10^6$ y and contributes to the long-term radiotoxicity. The cross section for population of both states is of primary interest for estimating the amount of long-term waste production when Bi is used in the cooling systems of nuclear reactors.

The value of the neutron-capture cross section for the isomeric state population was previously established as 17.7(7) mb [1], while the neutron-capture cross section leading to the ground state is more difficult to determine. Such studies rely significantly on a precise knowledge of the α_{320} conversion coefficient of the 320-keV, $2^- \rightarrow 1^-$ main ground-state transition. The value of α_{320} conversion coefficient cannot be calculated precisely, due to the fact that it depends on the $M1/E2$ multipolarity mixing of the 320-keV line, which so far has not been measured with sufficient precision. In previous studies, it was inferred from theoretical considerations that the 320-keV transition could be of almost pure $M1$ character [1]. However, this assumption has not been confirmed experimentally. In Ref. [1], the authors report three possible values of neutron-capture cross section to the ground state: 21.5(9), 19.3(8), and 17.2(7) mb, depending on the assumed multipolarity of the 320-keV γ transition, i.e., pure $M1$, 50% $M1$ + 50% $E2$, or pure $E2$, respectively.

We present revised calculations of the value of the neutron-capture cross section to the ground state in ^{210}Bi, with significantly reduced uncertainty. This was possible after defining with high accuracy the multipolarity mixing for the 320-keV line, as extracted from the γ angular correlation data collected with the HPGe EXILL array, at Institut Laue-Langevin in Grenoble (France). The analysis involved the minimization of the multivariable χ^2_Σ function

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constructed from the experimental angular correlation coefficients of 7 pairs of strong γ rays in ^{210}Bi.

2. Experiment and data analysis

2.1. Experimental setup

The cold-neutron capture reaction on ^{209}Bi was used to investigate the low-lying structure of the ^{210}Bi nucleus. The experiment was performed at the Institut Laue-Langevin (Grenoble, France) on the PF1B cold-neutron facility. After collimation, the capture flux on target was 10^8 neutrons/(s $\times$ cm^2). The EXILL array consists of 16 HPGe detectors: 8 EXOGAM clovers [2], 6 GASP detectors [3] and two clovers from the ILL LOHENGRIN instrument and has been used to measure coincidences between γ rays [4,5].

The collected data were sorted offline into a $\gamma\gamma$-coincidence matrix and a $\gamma\gamma\gamma$-coincidence cube with a time window of 200 ns. Based on the present data, the decay scheme of ^{210}Bi from the capture state (at 4605.2(1) keV) was established [6]. A large number of paths was found: 64 primary γ rays were identified, including 40 newly found branches. They feed the lower-lying states populating a complex level structure: a total of 70 discrete states were observed.

2.2. Angular correlations of γ rays

The 8 detectors of EXOGAM were arranged around the target each at 45°, forming a ring in a plane perpendicular with respect to the beam. This allowed us to sort double $\gamma\gamma$-coincidence data into three matrices corresponding to average angles between detectors of 0°, 45° and 90°. The analysis of γ-ray angular correlations provided information about transitions multipolarities, which confirmed previously known spins as well as helped with defining new assignments. We have focused on the determination of the 320-keV γ ray multipolarity, which has a significant impact on the calculations of the neutron-capture cross section to the ground state in ^{210}Bi.

The well-known formalism describing the anisotropy in the emission of γ rays with respect to the nuclear spin direction was applied [7,8]. The angular correlation function is usually expressed by the formula:

$$W(\theta) = A_0[1 + A_2 P_2(\cos\theta) + A_4 P_4(\cos\theta)], \quad (1)$$

where $P_m(\cos\theta)$ $(m = 2, 4)$ are Legendre polynomials, A_0 is the normalization coefficient, while values of A_m depend on the character of the two transitions considered. These parameters change with the δ_k mixing ratios ($k = 1, 2$ indicates the number of transition), which is the ratio of intensities of $L+1$ pole to L pole radiation [7]. Analysis of the non-stretched transition requires the minimization of the χ^2 cost function for $\delta_k \neq 0$:

$$\chi^2 = \left(\frac{A_2 - A_2^{theor}(\delta_k)}{\Delta A_2}\right)^2 + \left(\frac{A_4 - A_4^{theor}(\delta_k)}{\Delta A_4}\right)^2, \quad (2)$$

where the experimental A_m and theoretical A_m^{theor} coefficients are compared under a particular hypothesis for the spins and multipolarities, as a function of the δ_k mixing ratio. The minimum of the χ^2 function points to the most probable value of the δ_k parameter.

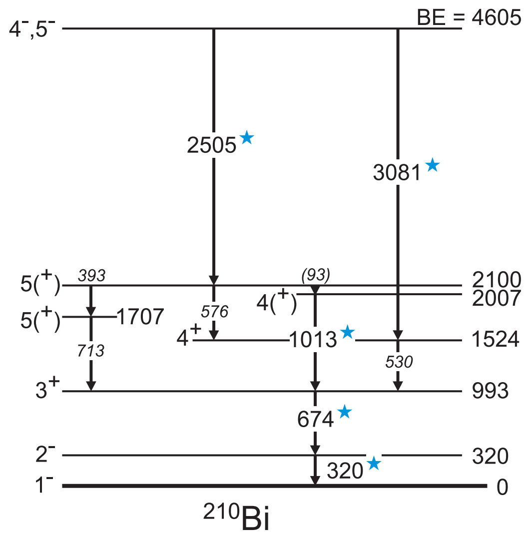

Figure 1. Partial decay scheme of ^{210}Bi produced in the cold-neutron capture reaction, showing the strongest decay paths leading to the ground state. The transitions used in the analysis are marked by asterisks. The full level scheme of ^{210}Bi is given in [6], as established from the present data.

The complete information about the admixture of higher order of multipole in a given γ ray can be obtained directly from angular correlations only if the second transition from the investigated pair is pure or its mixing ratio is firmly established. Knowledge of the transitions multipolarities is rather scarce, and there is no known γ ray with a firm multipolarity assignment in coincidence with the 320-keV line. For example, one can consider the 674-320-keV cascade leading to the ground state from the 993-keV level (Fig. 1). The angular correlation function constructed for this pair of transitions is displayed in Fig. 2(a) (red curve). The discrepancy between theoretical calculations performed for the hypothesis of pure $E1 - M1$ cascade (yellow dashed-dotted line in Fig. 2(a)) and the experimental result suggests multipolarity mixing in one or both transitions. However, to find the δ_k values in this case one must take into account other pairs of transitions being in coincidence with the 674-320-keV cascade, and use the multivariable minimization method outlined in the next section. A detailed description of this procedure can be found in [9].

2.3. Multivariable minimization

When the character of both γ rays in a cascade is not known, a minimum of 3 transitions, coincident with each other, is required in order to extract the mixing ratios by means of the angular correlation technique. We have found three very strong γ rays in coincidence with the 674-320-keV pair (i.e., the 1013-, 2505-, and 3081-keV lines). All five transitions are marked by asterisks in Fig. 1. They were combined into 7 pairs of γ rays in order to obtain 7 independent angular correlation functions. Next, the fitted A_{n2} and A_{n4} coefficients were used (Fig. 2) to construct the χ_n^2 functions with the formalism given by Eq. (2) (n = 1, ..., 7 indicates a given pair of γ rays). Each χ_n^2 function depends on two parameters, δ_{n1} and δ_{n2} as no value of mixing ratio is known (the mixing parameters are denoted later as e.g., δ_{320}, where the index refers to the

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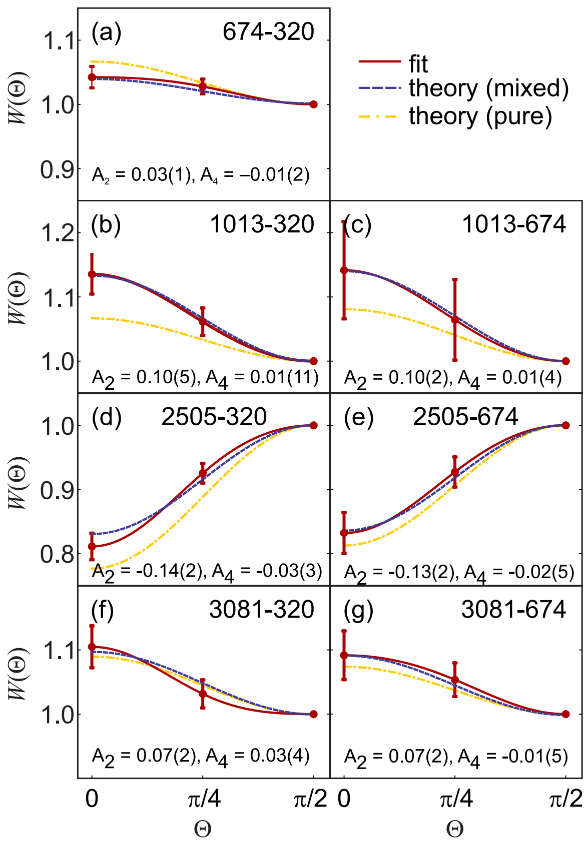

Figure 2. Seven considered angular correlations functions for ^{210}Bi. The functions fitted to the experimental points are marked by red solid lines, with the A_2 and A_4 coefficients given in the legend (see Eq. (1)). The theoretical curve calculated for the hypothesis of pure transitions is marked by the yellow dashed-dotted lines, while the blue dashed lines indicate the calculations including the mixing ratios extracted in the present analysis. As the value $W(90^\circ)$ was normalized to be 1, the error of the last point is included in the errors of the $W(0^\circ)$ and $W(45^\circ)$ points.

energy of the transition):

$$\chi_n^2 = \left(\frac{A_{n2} - A_{n2}^{theor}(\delta_{n1}, \delta_{n2})}{\Delta A_{n2}}\right)^2 + \left(\frac{A_{n4} - A_{n4}^{theor}(\delta_{n1}, \delta_{n2})}{\Delta A_{n4}}\right)^2 \quad (3)$$

An example of the single χ_n^2 function for the 674–320-keV pair of transitions is reported in Fig. 3(a). The χ_n^2 function does not have any well-defined minimum, so many $(\delta_{674}, \delta_{320}) \neq 0$ are possible in this case. Therefore, in order to define the δ_{320} mixing ratio, the cost function χ_Σ^2 was constructed in the following form:

$$\chi_\Sigma^2 = \frac{1}{\nu}\sum_n \chi_n^2, \quad (4)$$

where $1/\nu$ is the normalization factor and ν is the number of degrees of freedom.

The nonlinear least-square problem defined by Eq. (4) was solved by minimizing the cost function χ_Σ^2, using the Downhill Simplex algorithm (also known as Nelder-Mead method) [10]. The minimization algorithm determined 3 lowest minima, which gives three sets of mixing ratios

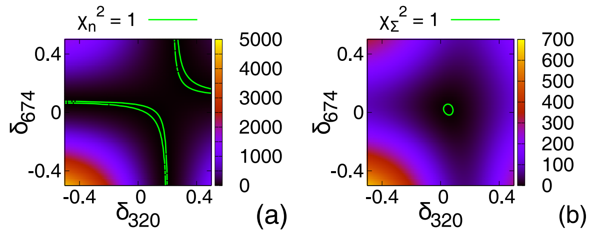

Figure 3. 2D projections of the (a) χ_n^2 and (b) χ_Σ^2 cost functions around the calculated minimum. Both functions are projected on the plane defined by the δ_{320} and δ_{674} mixing ratios.

for investigated transitions, with very similar values of χ_Σ^2 [9]. For those three minima, only two different values of δ_{320}: 0.05(2) and $-3.04(13)$ were found. The $\delta_{320} = -3.04$ value would imply a significant (90%) admixture of $E2$ multipolarity, which, in consequence, would result in a lower value of the neutron-capture cross section to the ground state in ^{210}Bi (comparing to this value calculated assuming pure $M1$ multipolarity of the 320-keV γ ray). However, as discussed in [9] with respect to the measurement of the half-life of the 320-keV state ($T_{1/2}$ = 7.5(14) ps [11]), one can conclude that the $\delta_{320} = -3.04$ solution is highly unlikely (the typical $T_{1/2}$ values for $E2$ in the neighboring nuclei would be much longer). Therefore, for the recalculation of the neutron-capture cross section to the ground state in ^{210}Bi, we adopted only the value δ_{320} = 0.05(2), which confirms almost pure $M1$ multipolarity of the 320-keV transition.

The quality of the minimization procedure is shown in Fig. 3(b) by projecting the χ_Σ^2 cost function on the plane defined by the δ_{674} and δ_{320} mixing parameters. The construction of the multivariable χ_Σ^2 results in a well pronounced minimum, in contrast to the single χ_n^2 function (as shown in Fig. 3, for the 674-320-keV pair of γ rays).

3. Recalculation of the neutron capture cross section

The value of $E2/M1$ mixing in the 320-keV transition can be then employed to recalculate the neutron-capture cross section to the ground state, σ_{gs}, in ^{210}Bi. By adopting the standard deviation of the extracted δ_{320} parameter, the 95% confidence range was calculated to be 0.024–0.076. We note that the presence of intermediate transitions in the investigated cascades may lead to an attenuation of γ-γ correlation, which would result in lower values of δ_{320} mixing parameter. Therefore, we consider δ_{320} = 0.076 as an upper limit and this value will be used to recalculate the neutron-capture cross section. This limit corresponds to the 0.6% admixture of $E2$ multipolarity in the 320-keV transition.

The cross section value can be obtained by following the analysis described in [12] and using the formula:

$$\sigma_{gs} = \frac{\sum_i I_i(1+\alpha_i)}{I_{4055}(1+\alpha_{4055})}\sigma_{4055}, \quad (5)$$

where $\sum_i I_i$ is the sum of the intensities of the γ rays leading to the ground state, reported by Borella *et al.* and corrected for internal conversion by the factor $(1+\alpha_i)$ (see Table 1). The σ_{gs} cross section was calculated relative to the partial capture cross section σ_{4055} = 8.07(14) mb

Table 1. The energies, assumed multipolarities and intensities of the γ rays (taken mainly from [12]) used for recalculation of the neutron-capture cross section are given in columns 1–3. Intensities marked with an asterisk come from the work [13]. Column four provides the correction factor for internal conversion.

E_γ	Multipolarity	I_γ [12]	(1+α)
320	M1+E2	1.721 (0.064)	1.388
348	E2	0.0268 (0.0017)	1.079
517*	(M1)	0.0133 (0.0066)	1.108
563	(M1)	0.0565 (0.0033)	1.086
972	(M1)	0.0565 (0.0033)	1.021
1118	(M1)	0.0129 (0.0014)	1.014
1175	(M1)	0.0371 (0.0031)	1.013
1531	(E1)	0.0032 (0.0029)	1.001
1585	(M1)	0.0086 (0.0013)	1.001
1981	(E1)	0.0048 (0.0031)	1.001
1990*	(E2)	0.0013 (0.0007)	1.002
2028	(E1)	0.0259 (0.0024)	1.001
4055	(M1)	1.0000 (0.0185)	1.002

for the very intense 4055-keV line in ^{210}Bi [12]. In this calculation the intensities of the 517- and 1990-keV γ rays, not observed by Borella *et al.*, were taken from [13]. The intensities of the 517-, 1118-, and 1981-keV γ rays were used to estimate the population of the 47-keV state feeding the ground state by a strongly converted $M1$ transition. The α_i conversion coefficients were obtained using the BrIcc calculator [14], assuming the lowest possible order of multipole for the transitions that do not have established multipolarities. The conversion coefficient for the line carrying most of the intensity, i.e., the 320-keV transition, was calculated taking into account its $E2/M1$ mixing ratio equal to 0.076. The resulting value of σ_{gs} is 21.3(9) mb. As the $\delta_{320} = 0.076$ should be considered as an upper limit, σ_{gs} was also calculated for pure $M1$ multipolarity of the 320-keV line to give a value of 21.5(9) mb.

4. Conclusions

We have proposed a recalculation of the neutron-capture cross section leading to ground state ^{210}Bi. $M1/E2$ multipolarity was adopted for the $2^- \rightarrow 1^-$ 320-keV line defined by an analysis based on minimization of a multivariable χ^2_Σ cost function [9] and the intensities reported in [12]. The $\delta_{320} = 0.076$, corresponding to an upper limit of 0.6% admixture of $E2$, defines the lower limit of σ_{gs}, i.e., 21.3 mb. The resulting range of possible σ_{gs}, 21.3(9)–21.5(9) mb, has been narrowed significantly, reducing the relative uncertainty on the ^{209}Bi(n,γ)^{210}Bi ground state cross-section from 25% [1] to 0.9%. These resulting cross section limits may serve for accurate projections of the ^{210}Po inventory in nuclear reactors and Accelerator Driven Systems when using Pb-Bi as coolant.

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