

Cross sections for formation of metastable states of ^{79}Br , ^{90}Zr , and ^{95}Tc nuclei at 14 MeV neutron energy

Ranjita Sarkar and V. N. Bhoraskar

Department of Physics, University of Poona, Pune 411007, India

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Cross sections for the formation of $^{79}\text{Br}^m$, $^{90}\text{Zr}^m$, and $^{95}\text{Tc}^m$ nuclear states respectively through the reactions $^{79}\text{Br}(n, n')^{79}\text{Br}^m$, $^{90}\text{Zr}(n, n')^{90}\text{Zr}^m$, and $^{96}\text{Ru}(n, n')^{95}\text{Tc}^m$ induced at 14 MeV neutron energy are measured in the laboratory using $^{27}\text{Al}(n, \alpha)^{24}\text{Na}$ as the reference reaction. The measured cross-section values (40 ± 3 mb for $^{79}\text{Br}^m$, 615 ± 28 mb for $^{90}\text{Zr}^m$, and 135 ± 8 mb for $^{95}\text{Tc}^m$) are in good agreement with the corresponding theoretical values (40 mb for $^{79}\text{Br}^m$, 584 mb for $^{90}\text{Zr}^m$, and 137 mb for $^{95}\text{Tc}^m$) estimated with compound nucleus theory and by assigning the isomer ratio in the limit 2 to 5 of the spin distribution factor. Probabilities for decay of excited nucleus to metastable and ground states via various possible spin states were calculated using a partial wave analysis method, an illustration of which is provided with an example of the reaction $^{115}\text{In}(n, 2n)^{114}\text{In}^m$. This method enables calculation of the isomer ratio even if the ground state of the nucleus is not radioactive.

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I. INTRODUCTION

Extensive theoretical and experimental work on cross sections of nuclear reactions induced by 14 MeV neutrons exist in the literature; however, very few results [1–5] are available on the formation of metastable states of nuclei. The importance of calculating isomeric cross-section values lies in the fact that they can be used to estimate various other nuclear parameters, such as the moment of inertia, level density, etc. Similarly, short-lived isomeric states may act as monitors to measure neutron flux both instantaneously or over a long period. Data on the formation of metastable states through 14-MeV neutron induced reactions are either not reported or scarce and in several cases remained unconfirmed without theoretical support [1–5]. A few workers adopted [1–5] a partial wave analysis approach to calculate isomeric cross sections for resonant and thermal-neutron induced reactions. Limited data exist on the estimation of isomeric cross sections [6] using nuclear spin distributions and Newton's level density formula, and on isomer ratios using Montalbetti's nomogram [7]. However, most of the results on isomeric cross sections are available for the cases in which both the metastable and ground states are radioactive. The present theoretical approach facilitates calculations of the isomeric cross section even if the ground state is not radioactive, and the nuclei of the present study $^{79}\text{Br}^m$, $^{90}\text{Zr}^m$, $^{95}\text{Tc}^m$, and $^{114}\text{In}^m$ fall under the same category. The partial wave analysis technique

was used to obtain probabilities for deexcitation of the excited states of ^{79}Br , ^{90}Zr , ^{95}Tc , and ^{114}In nuclei to different energy levels including metastable states. In several cases [5] cross sections were measured employing Van de Graaff machines where a deuterium ion beam at energies of 400 keV and above was used. In the d-t reaction, the neutron energies vary from 14.7 to 15.8 MeV for deuterium ion energies of 175 to 500 keV, respectively [8,9]. The cross-section value may therefore depend on the deuterium ion energy and, for this reason, the cross section of $^{90}\text{Zr}^m$ is remeasured in the present work.

II. EXPERIMENTAL PROCEDURE

The experimental procedure was similar to that described in an earlier publication [8]. Samples used were of enriched isotopes in powder form and the neutron generator facility [8] of this laboratory was used to carry out the present work. Neutrons were produced through the d-t reaction at 175-keV deuterium ion energy. Each sample powder of about 200 mg by weight was packed in a polyethylene vial, and, along with an aluminium foil, was irradiated with 14-MeV neutrons of flux $\approx 10^8$ /(cm²/sec). Both the aluminium foil and the sample were positioned almost at 0° with reference to the deuterium ion beam, enabling irradiation with monoenergetic neutrons. The sample after irradiation was transferred to a data collection room through a pneumatic transfer system and the induced gamma-ray activity

TABLE I. Details of the activation cycles and radioisotopes produced.

Reaction	Irradiation	Period of Cooling	Counting	Half-life of product	γ -ray energy	No. of cycles
$^{79}\text{Br}(n, n')^{79}\text{Br}^m$	5 sec	2 sec	5 sec	4.9 sec	0.21 MeV	400
$^{96}\text{Ru}(n, n')^{95}\text{Tc}^m$	1 h	10 min	1 h	61 days	0.204 MeV	5
$^{90}\text{Zr}(n, n')^{90}\text{Zr}^m$	2 sec	1 sec	2 sec	0.8 sec	2.32 MeV	400

TABLE II. A comparison of the presently estimated primary cross sections (in mb) with literature values.

Reaction	Experimental	Present theoretical
$^{69}\text{Ga}(n,p)^{69}\text{Zn}$	14.31 [16]	11.4
$^{64}\text{Zn}(n,n')^{64}\text{Zn}^*$	1358 [9]	1290
$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	$113.7 \pm .01$ [9]	123

was measured with a coaxial HPGe(38%) detector coupled to a multichannel analyzer. In the case of Zr and Br, only the sample was transferred after each irradiation to the detector, and after measurement of the gamma-ray activity was returned for reirradiation. However, the aluminium foil remained at its position for the total irradiation period of the sample. During activity measurement, the deuterium beam was deflected out of the tritium target. Four hundred activation cycles were repeated for Br and Zr and five for Ru. A standard computer program was used to calculate the area under each photopeak. Table I gives details about the activation cycle and gamma energies measured. The cross section σ for each nuclear reaction was calculated using the relation

$$\sigma = \frac{A\lambda}{N\phi\epsilon f_s f_d (1 - e^{-\lambda T})(e^{-\lambda t_a} - e^{-\lambda t_b})}$$

where A , λ , N , ϕ , f_s , f_d , T , t_a , t_b , ϵ , and σ are the activity, decay constant, number of the target atoms, neutron flux, self-absorption coefficient, branching ratio, period of irradiation, period of cooling, period of counting, efficiency of detector, and cross section, respectively. The neutron flux was estimated by measuring gamma-ray activity of ^{24}Na formed through the reaction $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$ for which the cross-section value 113.7 mb [9] is precisely known.

III. CALCULATIONS FOR ISOMERIC CROSS SECTIONS

A. Cross Section for compound nucleus and its deexcitation

At 14 MeV neutron energy, the cross section for formation of a compound nucleus [10] and subsequently the probability of its deexcitation through emission of a particle k , which can be a n , p , or α , were calculated, assuming that the probability for decay through γ emission is negligible. Pairing energy corrections, calculated using the Lang-Lecouteur [11] formula, were applied through the excitation energy [12] and level densities. In case of the (n, n') nuclear reaction, emission of the first neutron is considered only to the energy limit below which second

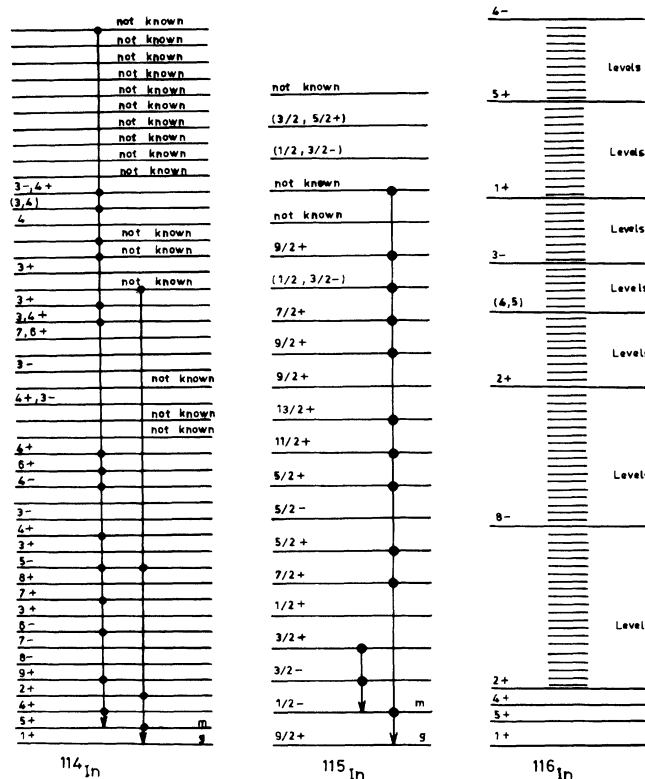


FIG. 1. Decay schemes of nuclei ^{114}In , ^{115}In , and ^{116}In involved in the reaction $^{115}\text{In}(n, 2n)^{114}\text{In}^m$.

neutron emission is energetically forbidden. Calculated values of cross sections for formation of excited states of ^{115}In , ^{96}Ru , ^{90}Zr , and ^{79}Br through the (n, n') reaction are 1871, 1556, 1653, and 1501 mb, respectively. To ensure that the present approach is correct, similar calculations for other elements were made and compared with the corresponding experimental values available in the literature, however results for only three nuclei are given in Table II. After first particle emission, the residual nucleus may have energy above threshold to emit a second particle, and with this approach cross-section values were calculated. The experimental cross-section value (1660 ± 166 mb) for the reaction $^{115}\text{In}(n, 2n)^{114}\text{In}$ is precisely known [9]. Our theoretical approach was tested by comparing the calculated value of the cross section, which was 1690 mb, Refs. [10–15] with this value. For further verification, cross sections for a few other reactions were also calculated with the same theory and were used to estimate cross sections for formation of metastable states (see Table III). The details of the calculations

TABLE III. A comparison of the presently estimated and measured values of cross sections (in mb) with the literature values.

Reaction	Literature		Present work	
	Expt.	Theor.	Expt.	Theor.
$^{74}\text{Se}(n, 2n)^{73}\text{Se}$		48.7 [17]	45 ± 3	40
$^{90}\text{Zr}(n, 2n)^{89}\text{Zr}$	600 [19]	677 [17]	604 ± 28	630
$^{96}\text{Ru}(n, n; p)^{95}\text{Tc}$	215 [20]	83.5 [20]		252

TABLE IV. Probability (P) for formation of a compound nucleus with spin J_f after emission of the first neutron.

J_f	$P(J_f)$
0.5	0.193 3609
1.5	0.790 5794
2.5	1.290 6500
3.5	1.916 2180
4.5	2.240 983
5.5	2.158 111
6.5	1.561 089

are given in steps, and have been illustrated with an example of the reaction $^{115}\text{In}(n,2n)^{114}\text{In}^m$. The decay schemes [15] of ^{114}In , ^{115}In , and ^{116}In with prominent γ -ray cascades are shown in Fig. 1.

B. Spin distribution of compound nucleus

The probability ρ for the formation of a compound nucleus with spin state J_c is given by [2]

$$\rho(J_c, E) = \pi \lambda^2 \sum_{|l=J_c-I-s|}^{|l=J_c+I+s|} \frac{2J_c+1}{(2s+1)(2I+1)} T_l(E), \quad (1)$$

where J_c and I are, respectively, spins of compound and target nuclei and similarly $T_l(E)$, E , s , and l are, respectively, the transmission coefficient, energy, spin, and angular momentum for the incoming particle. For all possible values of J_c , the probabilities for formation of the compound nucleus were obtained; however, in calculations only that value of J_c was used for which the probability was maximum. For the formation of $^{114}\text{In}^*$ (excited state) through the $^{115}\text{In}(n,2n)^{114}\text{In}^*$ reaction, the values of different parameters used in expression (1) are $E = 14$ MeV, $s = 0.5$, and $I = 5/2^+$, the minimum and maximum spin values are 1 and 9, respectively, and transmission coefficients $T_l(E)$ correspond to angular momenta l ranging from 1 to 14 [16]. The maximum probability (ρ) was obtained for J_c equal to 5, which was later on used in calculations of the residual nucleus spin distribution.

C. Spin distribution of the residual nucleus

Probabilities for transitions of a nucleus from its initial spin state J_c to a spin state J_f by emitting a particle are given by [2]

TABLE V. The probability values (P) for transition of the nucleus after particle emission to spin state J_f .

J_f	$P(J_f)$
1	0.500 4548
2	1.117 395
3	1.937 4
4	2.504 423
5	2.506 186
6	1.975 836
7	1.375 881
8	0.903 7365
9	0.462 7047

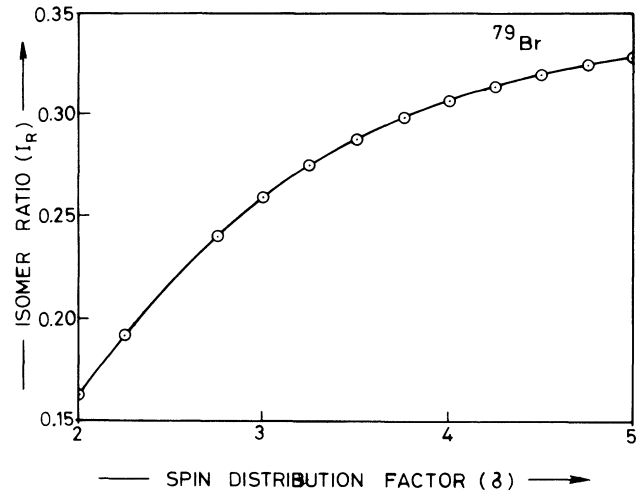


FIG. 2. Plot of isomeric ratio versus the spin distribution factor in the range 2 to 5 for the reactions $^{79}\text{Br}(n, n')^{79}\text{Br}^m$.

$$P(J_f) \propto \omega(J_f) \sum_{|l=J_c-J_f-s|}^{|l=J_c+J_f+s|} T_l(E), \quad (2)$$

where $\omega(J_f)$ is the level density factor given by the expression

$$\omega(J_f) \propto \omega(0)(2J_f+1)\exp[-(J_f+1/2)^2/2\delta^2]. \quad (3)$$

In the above expression $\omega(0)$ is the density of levels with spin zero, δ is the parameter which characterizes the distribution in spin, and J_c is the compound nucleus spin, for which the probability of decay is a maximum as calculated in the first step. Similar calculations are followed if emission of another particle is considered. To calculate the cross section for formation of $^{114}\text{In}^*$ (excited state), the values of different parameters considered are $E = 14$ MeV, $J_c = 5$, $J_f = 0.5$ to 6.5, and the angular momentum l ranges from 0 to 7. From the expression (2) the values of $P(J_f)$ were calculated by varying J_f in the range 0.5 to

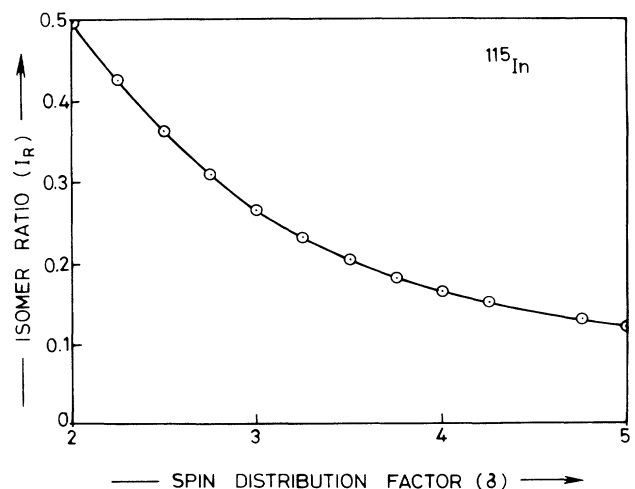


FIG. 3. Plot of isomeric ratio versus the spin distribution factor in the range 2 to 5 for the reaction $^{115}\text{In}(n, n')^{115}\text{In}^m$.

TABLE VI. Cross section and δ values for formation of a metastable state.

Reaction	Present work			Literature
	Theoretical	Experimental	δ value	
$^{79}\text{Br}(n, n')^{79}\text{Br}^m$	40.6 mb	$40 \pm$ mb	2	
$^{90}\text{Zr}(n, n')^{90}\text{Zr}^m$	584 mb	615 ± 28 mb	5	630 ± 31 mb [18]
$^{96}\text{Ru}(n, n')^{96}\text{Ru}^m$	137 mb	135 ± 8 mb	5	
$^{115}\text{In}(n, n')^{115}\text{In}^m$	62 mb		2.5 [9]	62 mb [9]

6.5, and the results are given in Table IV.

In the reaction $^{115}\text{In}(n, 2n)^{114}\text{In}^m$, following the emission of the first neutron, a second neutron is also emitted, probabilities $P(J_f)$ of which were calculated for all the J_f values in the range 0 to 9, results of which are given in Table V.

D. Metastable state formation

After emission of the second particle, the nucleus can go to any of the excited states defined by the quantum mechanical approach. Based on the decay schemes [15], different excited states of the residual nucleus were considered. The nucleus may decay directly to the metastable state or to any of the higher-energy states and then subsequently decay to ground state by emission of gamma rays. The probability for the decay of the excited nucleus to any of the levels was calculated using the spin value of each level. The parameter δ , given in Eq. (1), characterizes the distribution of spin. Theoretically, δ^2 is proportional to the product of the moment of inertia and nuclear temperature [1]. This indicates a dependence of δ on atomic number A of the nuclei. The probabilities for transition of the excited nuclei to metastable states σ_m and directly to ground state σ_g through different possible channels were added separately and the isomer ratio I_R was calculated using the relation

$$I_R = \frac{\sigma_m}{\sigma_g + \sigma_m} \quad (4)$$

IV. RESULTS AND DISCUSSIONS

Variations in the isomer ratio with the spin distribution factor for $^{79}\text{Br}^m$ and $^{115}\text{In}^m$ are shown respectively in Figs. 2 and 3. For $^{114}\text{In}^*$ the sum of the probabilities for

metastable state formation σ_m and that for ground state σ_g are 65.09 and 35.04, respectively. From the relation (4) the estimated value of the isomer ratio is 0.65, and therefore the cross section for the reaction $^{115}\text{In}(n, 2n)^{114}\text{In}^m$ is 1098 mb. The experimental value for the reaction is 1200 ± 120 mb [9] which is close to being within experimental error of the calculated value. Similar graphs were also plotted for $^{90}\text{Zr}^m$ and $^{95}\text{Tc}^m$, and the appropriate values of isomeric ratio for $^{79}\text{Br}^m$, $^{90}\text{Zr}^m$, $^{95}\text{Tc}^m$, and $^{115}\text{In}^m$ were found corresponding to δ values in the limit 2 to 5; these results are given in Table VI. This indicates that for both cases in which the isomer ratio increases or decreases with δ , the low atomic number in a particular range favors low value of δ and vice versa. This variation can also be observed in values reported by Eapen *et al.* [5]. Small differences in the experimental and theoretical results may be attributed to the unavailability of the spin values of the nuclear levels at higher energy [15], and therefore the contribution of the corresponding transition to different energy levels could not be estimated. Due to differences in the neutron energy the presently measured cross section for $^{90}\text{Zr}^m$ is slightly less compared to an earlier reported value shown in Table VI. In general all the values of cross sections calculated theoretically and given in Tables II, III, and VI are very close to corresponding to either present experimental or literature values. This indicates that, using the present approach, it is possible to calculate to fairly good accuracy cross sections for formation of metastable states of nuclei at 14 MeV neutron energy.

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