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Study on the Isomeric Ratio by Thermal Neutron Activation

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Abstract

The cross-section ratios of the nuclear isomeric pairs $^{80}Br^{m,g}$, $^{81}Se^{m,g}$, $^{104}Rh^{m,g}$, $^{116}In^{m,g}$ and $^{134}Cs^{m,g}$ through the radiative thermal neutron capture process have been studied. The experimental values of these ratios obtained by the activation method have been compared with the calculated ones deduced from the modified Huizenga-Vandenbosch method. Agreement between these values within 30% could be attained by controlling the spin cut-off parameter and gamma-ray multiplicity.

요 약

열중성자 포획 과정을 통한 핵이성체 쌍 즉 $^{80}Br^{m,g}$, $^{81}Se^{m,g}$, $^{104}Rh^{m,g}$, $^{116}In^{m,g}$ 및 $^{134}Cs^{m,g}$의 단면적 비를 다루었다. 방사화 방법에 의하여 구하여진 이들 핵이성체 쌍의 단면적 비의 실험측정치를 수정 보완된 Huizenga-Vandenbosch 법에 의한 계산치와 비교 검토하였다. 상기 측정치와 계산치는 spin cut-off parameter와 γ-선의 중복도를 적절히 조정함으로써 30% 이내의 오차 범위내에서 일치시킬 수 있었다.

1. Introduction

The theoretical estimates of the radiative transition probabilities between two nuclear states in de-excitation processes of a compound nucleus are practically impossible except the case of the deuteron formation because of the lack of the detailed knowledge for the formulation of the specific sets of the matrix elements which connect each possible state with the excitation energy E, the nuclear spin J, and the parity π[1, 2, 3]. However, during the last decade somewhat successful method of interpreting the cross-section ratios for the production of the nuclear isomeric pairs has been established using the statistical theory on the nuclear level density in conjunction with the Fermi gas model[4-14].

Huizenga and Vandenbosch[9, 10] proposed an approximate method for the calculation of the isomeric cross-section ratio

$$\eta=\frac{\sigma_H}{\sigma_H+\sigma_L}, \quad (1)$$

where σ_H and σ_L are the cross sections for the isomer production with high and low angular momentum, which are obtained by taking account of only the spin part in the level density formula assumed as follows:

$$\rho(E,J)\approx\rho(E)\cdot\frac{(2J+1)}{2\sigma^2}\cdot\frac{\exp\left(-\frac{J(J+1)}{2\sigma^2}\right)}{\sigma\sqrt{2\pi}}, \quad (2)$$

where the level density $\rho(E,J)$ is expressed in terms of the given excitation energy E, the angular momentum J and the spin cut-off parameter σ[15-17]. As an approximation, the transition probability from the state with the spin J to that with the spin J-1 or J or J+1 is calculated using the spin-dependent level density factors and the following relations:

$$P_{J\to J-1}=\frac{\rho(J-1)}{\rho(J-1)+\rho(J)+\rho(J+1)}$$
$$P_{J\to J}=\frac{\rho(J)}{\rho(J-1)+\rho(J)+\rho(J+1)} \quad (3)$$
$$P_{J\to J+1}=\frac{\rho(J+1)}{\rho(J-1)+\rho(J)+\rho(J+1).}$$

In this approximation, the underlying assumption is that the variations in the transition matrix elements were averaged out through a sufficiently large number of initial and final states. In the meanwhile, modified calculations with some improvements for the data analysis have been reported[3,10,14] but there are not yet the practically useful method to determine which parameter governs essentially the ratio between the isomeric level populations.

In the present paper, the spin distributions at the metastable or the ground state produced by emitting N_γ cascade dipole radiations from the compound initial state have been calculated for extensive values of parameters σ and N_γ. In the above calculation we used the same spin-dependent level density formula as done by Huizenga and Vandenbosch but excluding the zero-zero transitions of angular momentum in each cascade. The isomeric ratios deduced from the tabulations of these calculations shown in Table 3 can be compared with our recent measurements. Such extended tabulations are considered worthwhile as an intuitive guidance to visualize the gamma-ray cascade transitions with their spin dependent probabilities. Though the comparison between the calculated and the experimental data such as the above method seems somewhat formal, reasonably good agreements are noteworthy. In our measurements, the specpure materials of natural compositions were used for the targets of neutron activation and the neutron irradiations of these samples were carried out at TRIGA Mark Ⅱ reactor(250kW) at KAERI. Calibration of the detector system was carried out using standard sources, *i.e.* ^{241}Am, ^{57}Co, ^{203}Hg, ^{22}Na, ^{137}Cs, ^{54}Mn, ^{60}Co and ^{88}Y supplied from IAEA, Vienna, and the overall uncertainties of these were about 1%. The nuclear data[18,19] relevant to the present study are shown in Table 1. When most of the isomers at the metastable state decay to their ground state, two isomers have the relationship of parent and daughter, and the growth and decay of a daughter follows[27]

$$A_g=\frac{\lambda_g}{\lambda_g-\lambda_m}A_m^\circ(e^{-\lambda_m t}-e^{-\lambda_g t})+A_g^\circ e^{-\lambda_g t}$$
$$A_m=A_m^\circ e^{-\lambda_m t}, \quad (4)$$

where g and m refer to the ground and the metastable state, t cooling time, A and A° the activities after t and zero of time interval respectively, and λ decay constant. When the isomer of the metastable state decays only to the ground state, the isomeric cross-section ratio is given by[11,18,20]

Table 1. Summary of nuclear data

Reaction	Isomers	Half-life	Radiations detected	Detector used
$^{79}Br(n,\gamma)$	^{80}Br	17.6min	0.662+0.618 MeV γ	$3''\times3''$NaI(Tl)
	$^{80}Br^{m}$	4.98hr		
$^{80}Se(n,\gamma)$	^{81}Se	18.6min	0.27+0.29 MeVγ	$3''\times3''$NaI(Tl)
	$^{81}Se^{m}$	57min		
$^{103}Rh(n,\gamma)$	^{104}Rh	43sec	0.56 MeV γ, 2.5 MeV β	$2''\times2''$NaI(Tl)
	$^{104}Rh^{m}$	4.41min		$1\frac{1}{2}''\times\frac{1}{4}''$ Anthracene
$^{115}In(n,\gamma)$	^{116}In	14sec	1.27 MeV γ	$3''\times3''$NaI(Tl)
	$^{116}In^{m}$	54.0min	1.29 MeV γ	
$^{133}Cs(n,\gamma)$	^{134}Cs	2.05y	0.569+0.605 MeV γ	$3''\times3''$NaI(Tl)
	$^{134}Cs^{m}$	2.98hr	0.127 MeV γ	

$$\frac{\sigma_g}{\sigma_m}=\frac{1}{1-e^{-\lambda_g t}}\left\{\frac{A_g^{o}}{A_m^{o}}(1-e^{-\lambda_m t})-\frac{\lambda_g}{\lambda_m-\lambda_g}(e^{-\lambda_m t}-e^{-\lambda_g t})\right\}-1. \quad (5)$$

For irradiatipn periods short compared to $\frac{1}{\lambda}$, Eq. (5) reduces to

$$\frac{\sigma_g}{\sigma_m}=\frac{A_g^{o}t_{\frac{1}{2}}(g)}{A_m^{o}t_{\frac{1}{2}}(m)}, \quad (6)$$

where $t^{\frac{1}{2}}$ is the half-life.

Using Eqs. (5) and (6), the isomeric cross-section ratios calculated from the ratios between the activities at the end of the irradiation period were shown in Table 2.

2. Formalism

The theory on the nuclear level density based on the Fermi gas model was studied by H. A. Bethe[15]. Thereafter, C. Bloch studied the theory on the nuclear level density using the statistical theory based on the independent-particle model.

According to the independent-particle model, the simplest method calculating the nuclear level density is to consider that the level density of the single particle is continuous and to use the approximation such as a continuous integral. Therefore, the level density $\rho(E,M)$ is expressed as follows:[16,17]

$$\rho(E,M)=\rho(E)\cdot(\sigma\sqrt{2\pi})^{-1}\exp\left(-\frac{M^2}{2\sigma^2}\right), \quad (7)$$

where E is the total excitation energy, M the projective total angular momentum and

Table 2. Thermal and epi-cadmium yield ratios

Produced nucleus	η_{Exp}					
	Thermal neutron			Epi-Cd neutron		
	This Work	Others	Ref.	This Work	Others	Ref.
^{80}Br	0.21±0.01	0.25±0.05	28)	0.19±0.02	—	—
^{81}Se	0.14±0.02	0.115±0.012	11)	0.29±0.02	0.161±0.016	11)
^{104}Rh	0.12±0.02	0.071±0.007	11)	—	—	—
^{116}In	0.69±0.007	0.74±0.04	29)	—	—	—
^{134}Cs	0.058±0.004	0.086±0.013	11)	0.074±0.011	0.073±0.011	11)

σ represents the dispersion of this projective quantum number. In the case that the system consisted of nucleons rotates about an fixed axis, the rotational kinetic energy is given by:

$$E_{kin,\,rot}=\frac{\hbar^2M^2}{2I}. \tag{8}$$

As this energy is not contributed to the internal excitation of the system or the nucleus, the effective excitation energy has to be taken into account provided that the motion is similar to the rotational one of a rigid body.

$$E_{eff}=E-E_{kin,\,rot} \tag{9}$$

If we substitute Eq. (9) into the energy E of the standard equation of the energy level density $\rho(E)\equiv\rho(E,M=0)\propto\exp\left(\frac{E}{T}\right)$, then

$$\rho(E)\propto\exp\left(\frac{E}{T}\right)\exp\left(-\frac{\hbar^2M^2}{2IT}\right). \tag{10}$$

So

$$\rho(E,M)=\rho(E,M=0)\cdot\exp\left(-\frac{\hbar^2M^2}{2IT}\right). \tag{11}$$

From Eqs. (7) and (11),

$$\sigma^2=\frac{I}{\hbar^2}T \tag{12}$$

is given, where T is the temperature of the nucleus in energy units. Furthermore, σ is called as the spin cut-off parameter. And if the nuclear level density is described as a function of nuclear spin J, then

$$\rho(E,J)\equiv\rho(E,M=J)-\rho(E,M=J+1)$$
$$\approx\rho(E)\cdot\frac{(2J+1)}{2\sigma^2}\cdot\frac{\exp\left(-\frac{J(J+1)}{2\sigma^2}\right)}{\sigma\sqrt{2\pi}}, \tag{13}$$

where the energy-dependent level density $\rho(E)$ is used as a type of either $\exp(2\sqrt{aE})$ or $\exp\left(\frac{E}{T}\right)$ according to models. Also, the spin cut-off parameter σ depends upon the excitation energy E in the following way[14]:

$$\sigma^2=8.89\times10^{-2}\sqrt{aE}\,A^{\frac{2}{3}}, \tag{14}$$

where a is the level density factor proportional to the state density of the single particle near the Fermi energy.

The normalized spin distribution for the values of $J-1$, J and $J+1$ has been determined from the relative spin-dependent level density factor $\rho(J)$[15, 16]

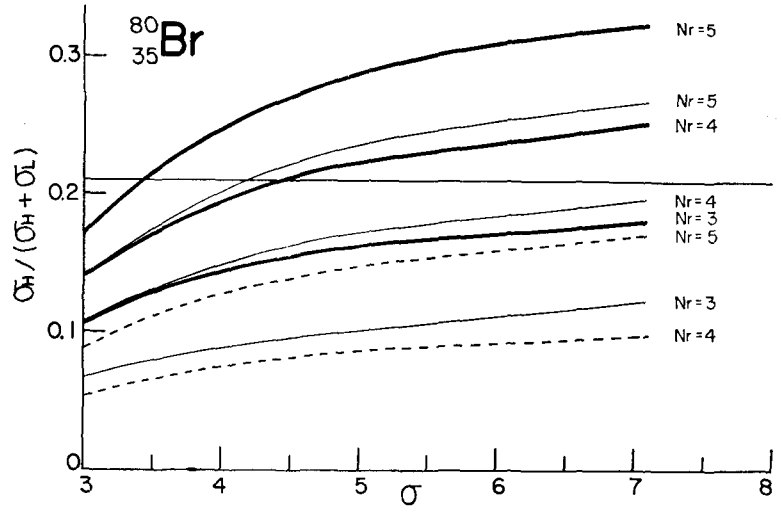
$$\rho(J)=\rho(0).(2J+1)\exp\left(-\frac{(J+\frac{1}{2})^2}{2\sigma^2}\right). \tag{15}$$

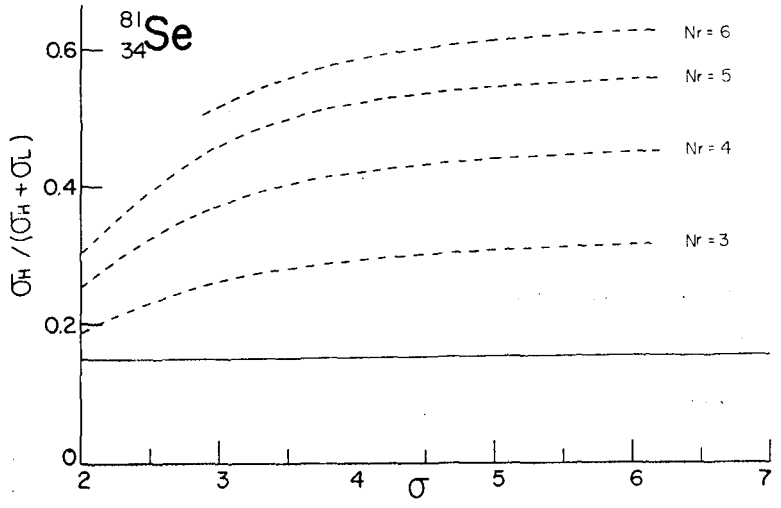
Taking account of the situation that the spin cut-off parameter σ is found to be often up to a value of 8[21] for an intermediate mass range, $\sigma=3,4,5$ and 7 in case of integer spins and $\sigma=2,3,4$ and 5 for half-integer spins are assigned repectively. Starting with an indicated initial state spin together with the conventional assumption that the gamma-ray cascade is consisted mostly of dipole radiation ($\Delta J=\pm1$ and 0) and using Eq. (3), the relative transition probabilities were calculated through cascades to reach at each final spin state after emission of $N\gamma$ dipole radiations. The zero-zero transitions of angular momentum which may be expected only through some internal processes are excluded in these calculations.

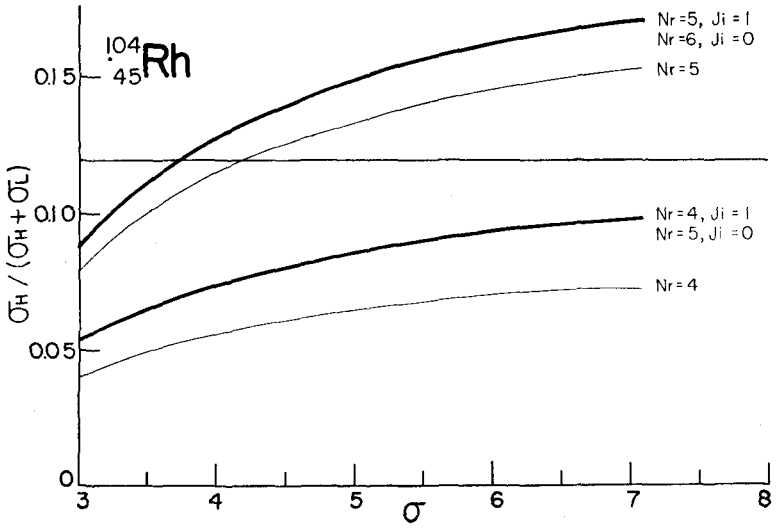
Finally in order to deduce any quantitative estimation on the isomeric cross-section ratio η from the calculated spin distributions, we assume the followings from the physical consideration.

(1) For thermal neutrons, as in case of the present experiment, only s-wave neutron capture will occur to give the spin of the capturing state in the compound nucleus by $I\pm\frac{1}{2}$, where I is the target nucleus spin.

(2) In general, the gamma-ray multiplicity has been kept as a significant variable for various calculations in nuclear spectroscopy. The average number of the gamma-ray cascade steps is believed approximately 2-5[3, 22, 23] and will tend to increase with mass number A[22-26]. Here a drastic choice of the value 5,


$^{80}_{35}$Br
Nr=5
Nr=5
Nr=4
Nr=4
Nr=3
Nr=5
Nr=3
Nr=4
$\sigma_H/(\sigma_H+\sigma_L)$
0.3
0.2
0.1
0
3
4
5
6
7
8
σ


$^{81}_{34}$Se
Nr = 6
Nr = 5
Nr = 4
Nr = 3
$\sigma_H/(\sigma_H+\sigma_L)$
0.6
0.4
0.2
0
2
3
4
5
6
7
σ


$^{104}_{45}$Rh
Nr=5, Ji = 1
Nr=6, Ji =0
Nr=5
Nr=4, Ji = 1
Nr=5, Ji =0
Nr=4
$\sigma_H/(\sigma_H+\sigma_L)$
0.15
0.10
0.05
0
3
4
5
6
7
8
σ

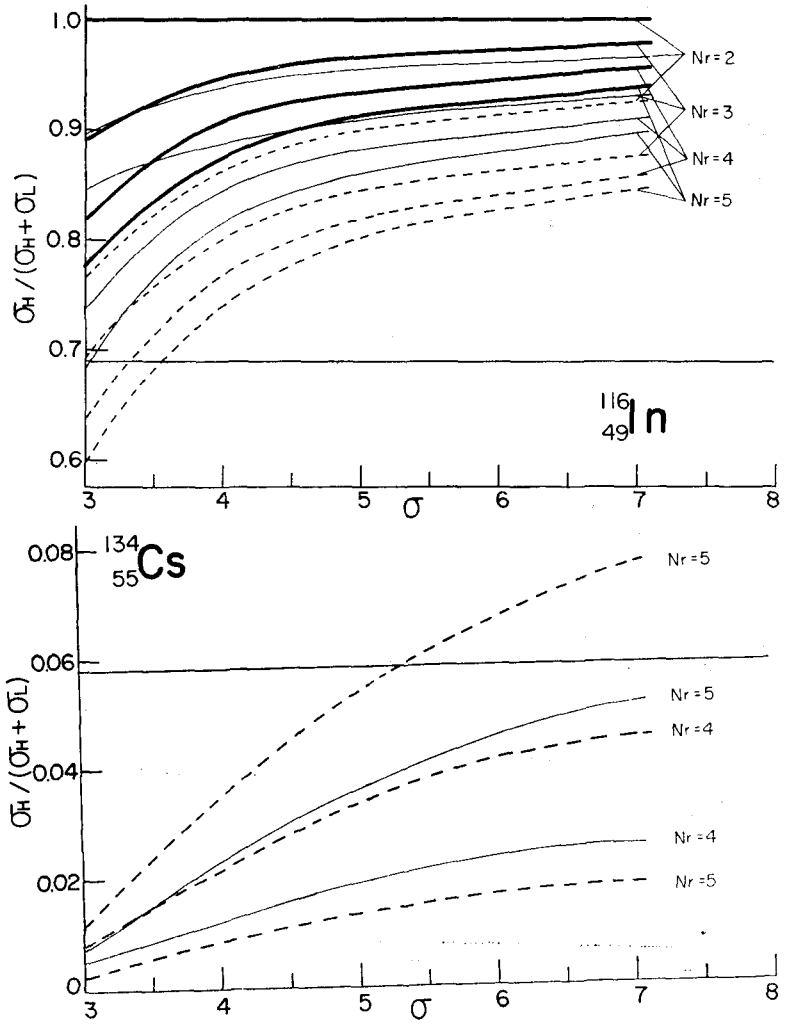


Figure 1. The isomeric cross-section ratios *vs.* the spin cut-off parameter are shown for the nuclei ^{80}Br, ^{81}Se, ^{104}Rh, ^{116}In and ^{134}Cs, respectively. The solid and dotted lines stand for the capture spin state of $J_i \pm \frac{1}{2}$, respectively. And the bold line stands for the total probability deduced from the summation of the above two states according to the (2J+1)−1aw.

except the value 3 for the case of ^{81}Se, has been adopted from a rough estimation by the formula, introduced by Nosov and Strutinsky on the basis of the statistical model[22],

$$N_r \approx \frac{1}{2}\sqrt{aE_r} \qquad (16)$$

(3) The third low-lying state decays into either of isomeric states by a last photon transition with the preference of lower multipolarity, as possible.

3. Conclusion and Discussion

The isomeric cross-section ratios η are shown in Figure 1 and in Table 3 for comparison with the present measurements. The

Table 3. Comparison of experimental and calculated isomeric cross-section ratios for thermal neutron capture

Nuclide formed in capture	Spin of capturing state	Spins of isomers*	η_{Exp}	η_{Calc}	Parameters
$^{80}_{35}Br$	2−	5, 1+(2)	0.21±0.01	0.171(0.247)	N_γ=5, σ=3(4)
	1−			0.088(0.128)	N_γ=5, σ=3(4)
$^{81}_{34}Se$	$\frac{1}{2}$	$\frac{7}{2}$, $\frac{1}{2}$−	0.14±0.02	0.261(0.184)	N_γ=3, σ=3(2)
$^{104}_{45}Rh$	1−	5−, 1+(2)	0.12±0.02	0.088(0.128)	N_γ=5, σ=3(4)
	0−			0.053(0.075)	N_γ=5, σ=3(4)
$^{116}_{49}In$	5+	5+, 1+	0.69±0.07	0.755(0.875)	N_γ=5, σ=3(4)
	4+			0.595(0.746)	N_γ=5, σ=3(4)
$^{134}_{55}Cs$	4+	8−, 4+(5)	0.058±0.004	0.034(0.053)	N_γ=5, σ=4(5)
	3+			0.008(0.013)	N_γ=5, σ=4(5)

* Competing level spin is given in parentheses.

phenomenal agreement between the theory and the experimental values within 30% has been experienced by the formal control of the parametric values within the reasonable numerical ranges for the prefered case of odd-odd nuclei, for which the residual nucleon interaction can be neglected in this mass range.

First of all, a primarily significant parameter must be determined in deducing the information on the isomeric pair formation as mentioned before. Thus far N_γ seems to be a most promising factor and it is much desirous to visualize the distribution of the number of photon transitions for a certain de-excitation process. Many studies have been done on nuclear decay schemes by various types of the nuclear reaction, and the practical significance of the determination of N_γ has been also emphasized by many authors[26].

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