

Formation of high-spin neodymium and mercury isomers in neutron and charged particle induced nuclear reactions

S. Sudár^{1,2,a}, K. Hilgers², M. Al-Abyad^{2,3}, and S.M. Qaim²

¹ Institute of Experimental Physics, University of Debrecen, Bem tér 18/a, 4026 Debrecen, Hungary

² Institut für Nuklearchemie, Forschungszentrum Jülich GmbH, 52425 Jülich, Germany

³ Cyclotron Facility, Nuclear Research Centre, Atomic Energy Authority, Cairo 13759, Egypt

Abstract. The isomeric pairs $^{195\text{m.g}}\text{Hg}$, $^{197\text{m.g}}\text{Hg}$, $^{139\text{m.g}}\text{Nd}$ and $^{141\text{m.g}}\text{Nd}$ constitute interesting cases since the ground state has a low spin and the isomeric state a relatively high spin. Their formation was studied in several reactions induced by n, p, ^3He , and α -particles. The four projectiles were produced at the Jülich variable-energy compact cyclotron (CV 28); for neutron production the $^2\text{H}(\text{d},\text{n})^3\text{He}$ reaction was used. The cross sections were measured by the activation technique. From the available experimental data, isomeric cross-section ratios were determined. Nuclear model calculations using the code STAPRE, which employs the Hauser-Feshbach and exciton model formalisms, were undertaken to describe the formation of both the isomeric and the ground states of the products. The calculations were compared with the results of the EMPIRE-II code. The total reaction cross section of a particular channel is reproduced fairly well by the model calculations, with STAPRE giving slightly better results. Regarding the isomeric cross sections, the agreement between the experiment and theory is only in approximate terms. A description of the isomeric cross-section ratio by the model was possible only with a very low value of η ($\eta = \Theta_{\text{eff}}/\Theta_{\text{rig}}$) for the mercury isotopes, while slightly larger η values were needed for neodymium isotopes. An exponential mass dependence of η is proposed.

1 Introduction

Studies of excitation functions of nuclear reactions are important for testing nuclear models and for practical applications. Furthermore, isomeric cross-section ratios are of fundamental significance. The isomeric pairs $^{195}\text{Hg}^{\text{m.g}}$, $^{197}\text{Hg}^{\text{m.g}}$, $^{139}\text{Nd}^{\text{m.g}}$ and $^{141}\text{Nd}^{\text{m.g}}$ appeared to be very interesting: in all cases the ground state has a low spin ($1/2^-$), ($1/2^-$), ($3/2^+$), ($3/2^+$) and the metastable state a higher spin ($13/2^+$), ($13/2^+$), ($15/2^-$), ($11/2^-$), respectively. In addition to commonly used projectiles, neutrons and protons, we also investigated reactions induced by ^3He - and alpha particles. The higher angular momentum brought by those projectiles might shed some more light on the effect of spin distribution of the level density on the isomeric cross-section ratio.

Using the framework of the Fermi gas model for the global parameterization of the nuclear level density, the spin distribution is described by the formula

$$\frac{\rho_J}{\rho} = \frac{2J+1}{2\sqrt{2\pi}\sigma^3} e^{-\frac{J(J+1)}{2\sigma^2}} \quad (1)$$

where ρ is the total level density, while ρ_J is the density of spin- J levels without the $2J+1$ degeneracy factor. The parameter σ is known as spin-cutoff parameter. The ρ , ρ_J and σ are all functions of the excitation energy.

In the empirical parameterization, the σ is determined using the rigid body moment of inertia (Θ_{rig})

$$\Theta_{\text{rig}} = \frac{2}{5}mA(r_0A^{1/3})^2 \quad (2)$$

^a Presenting author, e-mail: sudarsa@delphin.unideb.hu

where r_0 is the nuclear radius parameter, A the mass number and m is the nucleon mass. Introducing the $\eta = \Theta_{\text{eff}}/\Theta_{\text{rig}}$, the square of the spin-cutoff parameter can be expressed as

$$\sigma^2 = \eta\Theta_{\text{rig}} \frac{T}{\hbar^2} \quad (3)$$

where T is the temperature in thermal ensembles.

Since Bethe's pioneering work [1] the nuclear level density problem has remained an active area of both theoretical and experimental studies. One way of studying the spin distribution of the levels is to measure the isomeric cross-section ratio as a function of the projectile energy.

2 Experimental

Cross sections were measured by the activation method, which is almost ideal for studying closely spaced low-lying isomeric states, provided their lifetimes are not too short. In work on charged particle induced reactions, high purity thin platinum and gold foils were stacked together. In the case of Ce and Pr targets, thin CeO_2 or Pr_2O_3 layer was prepared by sedimentation on Cu foil. In a given stack about 4 or 5 such foils were placed together with the monitor foils, which also served to degrade the projectile energy as well as to determine the incident particle energy and the beam intensity. Each stack was irradiated for 30–60 minutes at the Compact Cyclotron CV 28 at the Forschungszentrum (FZ) Jülich. The primary energies used were 27.7 MeV for α -, 36.9 MeV for ^3He -particles and 20 MeV and 45 MeV for protons. Protons of energy 45 MeV were extracted from the injector of COSY. The average particle energy effective at each foil was calculated

using the standard formalism and corrected by the method described in [2]. The energy degradation along the stack and the beam current were checked by the reactions induced in the inserted monitor foils. In work with neutrons, pellets of HgCl_2 with monitor foils were irradiated using D+d neutrons at the cyclotron.

A HPGe and a low energy HPGe detector were used to measure the activities of the irradiated monitor foils and the activities of $^{195}\text{Hg}^{\text{m,g}}$, $^{197}\text{Hg}^{\text{m,g}}$, $^{139}\text{Nd}^{\text{m,g}}$ and $^{141}\text{Nd}^{\text{m,g}}$ in the irradiated samples. Peak area analyses were done using the PC version of the GAMANAL spectrum analyser programme. The detector efficiency was determined experimentally using a selected set of γ -ray standard sources. The true coincidence corrections for the gamma lines were calculated by the TrueCoinc code. The details have already been reported [3–5].

The count rates at the end of bombardment (EOB) were converted to decay rates A_g , A_m by introducing corrections for emission probabilities of γ -rays, detector efficiency, self-absorption, coincidence loss, dead time, and random pile up. Cross sections were calculated using the activation equation. The isomer ratio was obtained from the ratio of σ_m to σ_g or σ_m to $\sigma_{\text{m+g}}$, depending on the experiment.

The individual uncertainties of the cross sections and cross section ratios were combined in quadrature, taking into account the correlation of the data, to obtain an overall uncertainty of 9–31% and 9–50% for the cross sections and for the isomeric cross-section ratios, respectively.

3 Nuclear model calculations

Cross sections were calculated using the statistical model taking into account the pre-equilibrium effects. In general, two codes, namely STAPRE and EMPIRE, were used.

3.1 STAPRE calculations

The details of the calculations are given elsewhere [3,4]. In the present work the emphasis was on the isomeric cross sections. Since such calculations are strongly dependent on the input level scheme of the product nucleus [6], we chose those parameters carefully. The energies, spins, parities and branching ratios of discrete levels were extracted by using the NNDC On-Line Data Service from the ENSDF database. In the continuum region the level density was calculated by the back-shifted formula with adjusted level density parameter. We characterized the spin distribution of the level density by the parameter η (see above) and the calculations were performed for different η values to find the best value getting agreement with the experimental data.

3.2 EMPIRE calculations

In most of the STAPRE calculations done in this work an η value of about 0.25 was found to be most suitable as compared to about 0.5 normally used. In order to check the credibility of those calculations we also used the EMPIRE-II (version 2.19) code [7]. In this case the standard library of input parameters

was used. For the proton induced reaction the direct contribution was determined via the Coupled Channel calculation using the built in ECIS03 code. The particle transmission coefficients were generated via the spherical optical model using the computer code (ECIS03) and the default set of global parameters. In the calculation the Multi Step Direct, Multi Step Compound, Hauser-Feshbach model with width fluctuation correction (HRTW), the DEGAS and PCROSS codes were used. For the level densities, the dynamic approach of the EMPIRE-II and HF-BCS microscopic level densities were used.

4 Results and discussion

Experimental and theoretical studies showed that the excitation functions of ^3He -, α -particle, proton and neutron induced reactions leading to the formation of the isomeric pairs $^{195}\text{Hg}^{\text{m,g}}$ and $^{197}\text{Hg}^{\text{m,g}}$ are described to varying degree of success by the statistical theory incorporating pre-compound effects.

Despite the relatively high spins of the metastable states, the isomeric cross-section ratios are relatively high for the two pairs even at low excitation energies; they increase with the increasing projectile energy. Detailed results have been already published [3,4]. Here we give typical results for the pair $^{197}\text{Hg}^{\text{m,g}}$ in three reactions in figures 1 to 3 and those for the $^{195}\text{Hg}^{\text{m,g}}$ in one reaction in figure 4. The isomeric cross-section ratio for each of the two pairs appears to be reproduced by the STAPRE calculation only when an η value of 0.25 or less is used. The ratios cannot be described by a nuclear reaction model code using the conventional level density formalism with the usual η value. The agreement between the STAPRE calculation with $\eta = 0.25$ and EMPIRE with HF-BCS microscopic treatment implies that the low η values originate from the microscopic structure of these nuclei.

The advantage of using the alpha and helium-3 induced reactions for studying the isomeric cross-section ratio is that the compound system gets larger angular momentum transferred than in nucleon-induced reactions. As a result, the isomeric cross-section ratio becomes more sensitive to the spin distribution.

The energy dependence of the isomeric cross-section ratio is described well by the model calculations. This implies

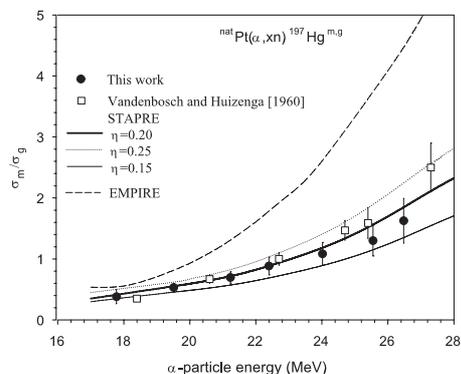


Fig. 1. Isomer ratio in the $^{\text{nat}}\text{Pt}(\alpha, xn)^{197}\text{Hg}^{\text{m,g}}$ process; for details see [3].

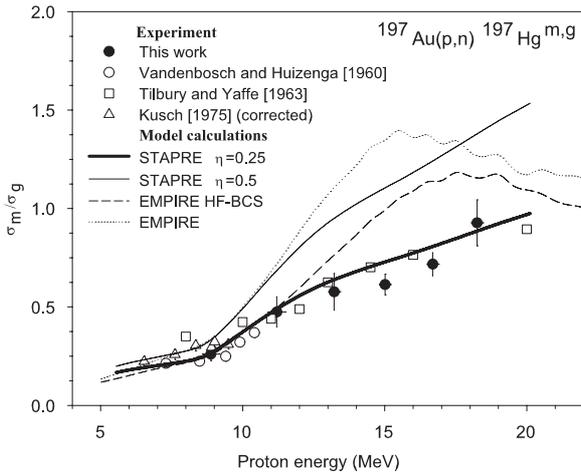


Fig. 2. Isomer ratio in the $^{197}\text{Au}(p,n)^{197}\text{Hg}^{m,g}$ process; for details see [3].

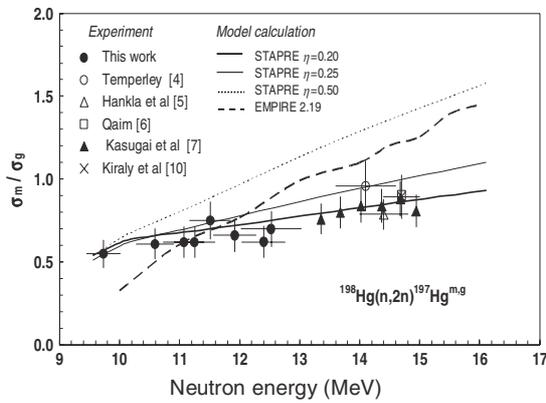


Fig. 3. Isomer ratio in the $^{198}\text{Hg}(n,2n)^{197}\text{Hg}^{m,g}$ process; for details see [4].

that the contribution of two proton stripping direct process is negligible. It also indicates that to a first approximation η is independent of energy and the energy dependence of the spin-cutoff factor is adequate. Similar results were obtained from measurements on neutron and proton induced reactions. Analyses of those measurements confirmed the low value of η .

The characteristics of the discrete levels as well as their branching ratios play an important role in the calculation of isomeric cross-section ratios. The constancy in the η value in describing (within the uncertainty of the experimental data) the different particle induced reactions (α , ^3He , p, n) shows that there cannot be any serious error in the decay scheme of the discrete levels.

Since the population of the discrete levels by particle emission and gamma cascade from the continuum will be different for different types of reactions, the error in the level scheme would lead to a large deviation between the needed η values to fit the calculated and measured isomeric cross-section ratios.

For example the maximum of the spin distribution for the ^{198}Hg compound nucleus is at $5\hbar$ in the $^{197}\text{Au}(p,n)^{197}\text{Hg}^{m,g}$

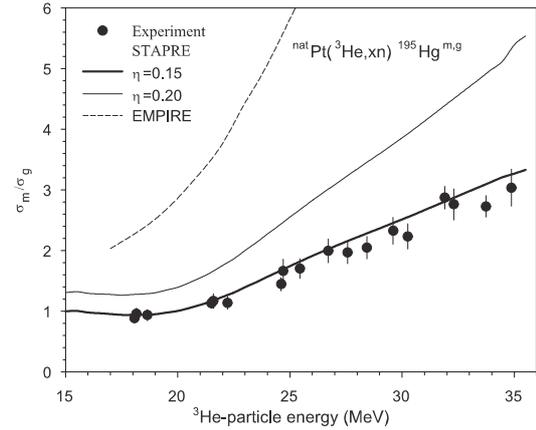


Fig. 4. Isomer ratio in the $^{\text{nat}}\text{Pt}(^3\text{He},xn)^{195}\text{Hg}^{m,g}$ process; for details see [3].

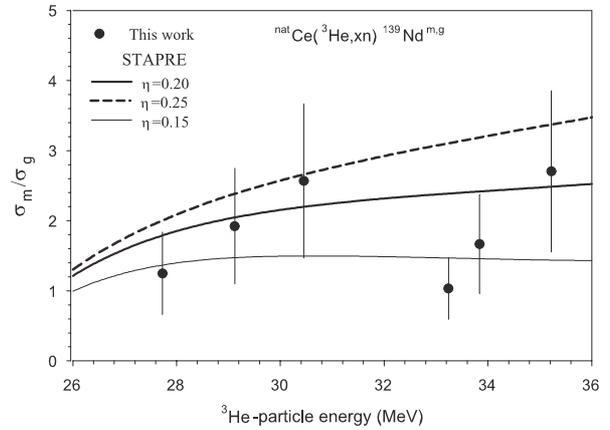


Fig. 5. Isomer ratio in the $^{\text{nat}}\text{Ce}(^3\text{He},xn)^{139}\text{Nd}^{m,g}$ process.

reaction but it is at $16\hbar$ in the $^{194}\text{Pt}(\alpha,n)^{197}\text{Hg}^{m,g}$ reaction at about the same excitation energy of the ^{198}Hg .

It may be argued that the spin-cutoff factor and the ratio of the effective moment of inertia Θ_{eff} to the rigid-body moment of inertia Θ_{rig} ($\eta = \Theta_{\text{eff}}/\Theta_{\text{rig}}$) show anomaly for the isotopes ^{195}Hg and ^{197}Hg . In order to dispel this doubt we performed further measurements on Nd isotopes.

The results for the pair $^{139}\text{Nd}^{m,g}$ are presented in figures 5 and 6. The best fits to the experimental isomeric cross-section ratios need η values of 0.20 and 0.24 in the $^{\text{nat}}\text{Ce}(^3\text{He},xn)^{139}\text{Nd}^{m,g}$ and $^{141}\text{Pr}(p,3n)^{139}\text{Nd}^{m,g}$ processes, respectively. Figure 6 also shows some recent experimental data on the $^{141}\text{Pr}(p,3n)^{139}\text{Nd}^{m,g}$ process reported by Steyn et al. [8].

These results confirm that the low value of η is not unique for the mercury isotopes. The results for the pair $^{141}\text{Nd}^{m,g}$ are not presented, since in this case the calculations were not sensitive enough to make any decision on the basis of the experimental data. The reason of it may lie in the smaller momentum difference and relatively low excitation energy.

In a recent work [9] the spin cutoff factor (σ) was evaluated at low excitation energy in the mass range of $20 \leq A \leq 110$. It was concluded, that using the formula

$$\sigma = \text{const}(U - \Delta)^{0.25} A^{5/6} / a^{1/4} \quad (4)$$

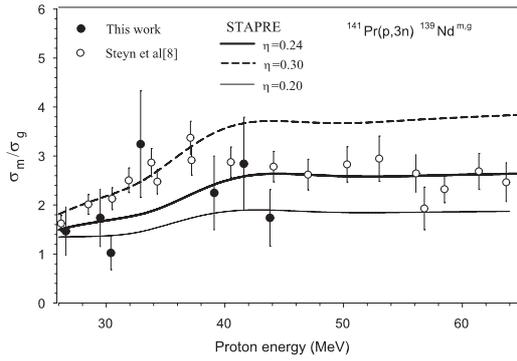


Fig. 6. Isomer ratio in the $^{141}\text{Pr}(p,3n)^{139}\text{Nd}^{m,g}$ process.

for the energy (U), energy shift (Δ), mass number (A) and level density parameter (a) dependence of σ the fitted “const” in the formula shows a mass number dependence: $\text{const} = 0.10578 - 0.000202A$.

This “const” is directly connected to the parameter η used in this paper and we converted the ref [9] data to η using the relation

$$\eta^{1/2} = \frac{\text{const}}{\text{const}_{\text{rigid}}}. \quad (5)$$

The results are plotted in figure 7, together with the results from our data and the recent analysis of ref. [10] suggesting $\eta = 0.75$ for ^{51}V . It can be seen from the evaluation of [9], that the individual values are strongly scattered around the trend shown in figure 7 by the dotted line, calculated as the average η for 10 mass numbers.

A lot of successful model calculations performed with $\eta = 0.5$ in the medium mass region, and our conclusion of $\eta = 0.22$ for $A = 139$, $\eta = 0.20$ for $A = 197$, and $\eta = 0.15$ for $A = 195$ fit well into the trend. The general decreasing tendency of the data seems to be exponential.

The fitted equation to all data is:

$$\eta = a \cdot e^{-bA}. \quad (6)$$

The best fit was obtained by $a = 0.6643$ and $b = 0.0065$.

These empirical results should be confirmed for the high mass number region with further experiments, and an explanation of mass dependence of η needs further theoretical work.

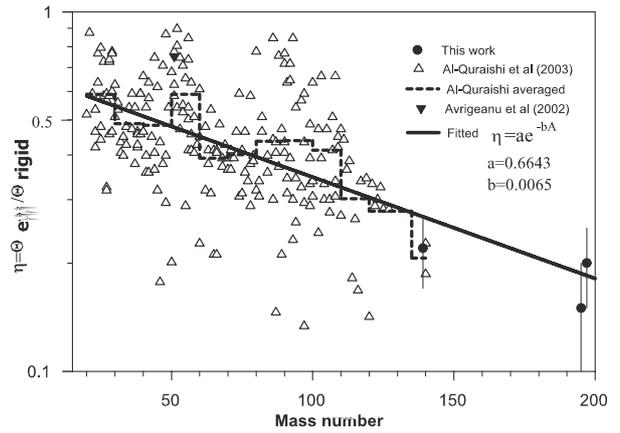


Fig. 7. Experimental values of $\eta = \theta_{\text{eff}}/\theta_{\text{rigid}}$ as a function of the mass number.

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References

1. H. Bethe, Phys. Rev. **50**, 332 (1936).
2. S. Sudár, S.M. Qaim, Phys. Rev. C **50**, 2408 (1994).
3. S. Sudár, S.M. Qaim, Phys. Rev. C **73**, 034613 (2006).
4. M. Al-Abyad, S. Sudár, M.N.H. Comsan, S.M. Qaim, Phys. Rev. C **73**, 064608 (2006).
5. K. Hilgers, Yu.N. Shubin, H.H. Coenen, S.M. Qaim, Radiochim. Acta **93**, 553 (2005).
6. S.M. Qaim, A. Mushtaq, M. Uhl, Phys. Rev. C **38**, 645 (1988).
7. M. Herman, R. Capote, B. Carlson, P. Oblozinsky, M. Sin, A. Trakov, V. Zerkin, *EMPIRE-II, Nuclear reaction model code, version 2.19* (International Atomic Energy Agency, Vienna, 2005).
8. G.F. Steyn, C. Vermeulen, F.M. Nortier, F. Szelecsényi, Z. Kovács, S.M. Qaim, Nucl. Instrum. Meth. B **252**, 149 (2006).
9. S.I. Al-Quraishi, S.M. Grimes, T.N. Massey, D.A. Resler, Phys. Rev. C **67**, 015803 (2003).
10. V. Avriganu, T. Glodariu, A. Plompen, H. Weigmann, *Proc. Int. Conf. on Nuclear Data for Science and Technology, Tsukuba, Japan, October 2001*, J. Nucl. Sci. Technol. Suppl. **2**, 746 (2002).