The Existence of Different Capture Levels in the Formation of Nuclear Isomers by Slow Neutron Irradiation

P. C. CAPRON AND A. J. VERHOEVE-STOKKINK*
Institut Inter-Universitaire de Physique Nucléaire, University of Louvain, Louvain, Belgium
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Experiments have been performed on the formation of bromine and rhodium isomers, by thermal as well as resonance neutrons, in order to study capture levels. With slow neutrons of variable energies obtained from a Ra—Be source in a slowing-down medium of paraffin, we have studied: (a) the relative formation of the isomeric pairs as a function of the distance from the source and (b) the migration distances for each isomer.

The experimental results show that for bromine isomers produced by resonance neutrons as well as for rhodium isomers produced by resonance and *C*-neutrons, the ratio of the activities of the fundamental state to the metastable state is a function of the distance from the source. Moreover, at each point this ratio is different for resonance and for *C*-neutrons. The migration distances corresponding to each of the isomers formed by resonance neutrons on bromine, or by *C*-neutrons on rhodium, are found to be different.

This leads to the assumption of the existence of more than one capture level in the formation of the bromine and rhodium isomers. It may be suggested that for rhodium the resonance levels extend within the region defined by the *C*-neutrons.

I. INTRODUCTION

THE question of whether nuclear isomers, such as the isomers of Br^{80}, Rh^{104}, or In^{119} arise from one or more capture levels, is still undecided.[1-9] In order to elucidate this problem, we have investigated the formation of two of these isomeric pairs by means of slow neutrons irradiation by the reactions

$$\begin{array}{l} \qquad\qquad\quad Br^{80}(4.4\text{ hr}) \\ \qquad\qquad\quad\nearrow \\ Br^{79}(n,\gamma)Br^{80*} \\ \qquad\qquad\quad\searrow \\ \qquad\qquad\quad Br^{80}(18\text{ min}) \end{array} \quad ,$$

$$\begin{array}{l} \qquad\qquad\quad Rh^{104}(4.2\text{ min}) \\ \qquad\qquad\quad\nearrow \\ Rh^{103}(n,\gamma)Rh^{104*} \\ \qquad\qquad\quad\searrow \\ \qquad\qquad\quad Rh^{104}(44\text{ sec}) \end{array} \quad .$$

The object was to examine separately "resonance" and "thermal" capture levels for both isomers and to work with a sufficiently high precision to decide whether neutron capture levels corresponding to nuclear isomers are simple or multiple.

Using a Ra—Be neutron source in a slowing-down medium of paraffin, we studied: (a) the relative formation of the isomeric pairs as a function of decreasing neutron energy (increasing distance from the source), (b) the migration distances of the neutrons in the paraffin for each isomer.

A variable ratio for the activities of the two isomers, as well as a difference between their migration distances, would give an indication in favor of the multiplicity of the capture levels.

II. PROCEDURE

The slow neutrons were produced by a Ra—Be neutron source fixed in the center of a paraffin block 50 cm high and 40 cm in diameter. The bromine or rhodium detectors were placed in the paraffin block at increasing distances from the source.

In one part of the experiments the detectors were wrapped in a Cd shield, so that activation was only due to "resonance" neutrons; *viz.* all the effective neutrons had energies higher than the Cd resonance energy (0.17 ev). The second part concerned experiments without Cd, the difference of the two series giving the activation of "*C*-neutrons," which are considered practically to be the neutrons in the thermal region (0.028 ev).

The bromine sample was irradiated for 15 hours by a one-Curie Ra—Be source. The detector consisted of a square, 3 cm per side, of carbon tetrabromide, 0.2 g/cm² wrapped in Cellophane.

Activity[10] due to the Br^{82} (34 hr) was eliminated, so that the remaining activity corresponded to the isomers Br^{80}(4.4 hr) and Br^{80}(18 min).

The rhodium sample was irradiated for 70 min. The activity caused by the isomer of 44-sec period is nearly ten times as high as that of the 4.2-min period, so that when we take the activity of the latter sufficiently high to be counted with good accuracy, the activity of the short-lived isomer is so high that the corrections for the resolving time of the counting apparatus would preclude

* Research physicist of the Laboratoire de Recherches Physiques des Ateliers de Constructions Electriques de Charleroi (A.C.E.C.), at Charleroi, Belgium.

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good accuracy. We found, indeed, that the resolving time may vary with the activity, as has already been stated by others.[11] Thus, in a first series of experiments we measured only the activity of the 4.2-min period as a function of the distance from the source. We therefore used a one-Curie source and a square metallic Rh sample 2.4 cm per side, of 0.123 g/cm².

In a second series of experiments we determined only the ratio of the activities of the two isomers, as a function of the distance from the source. This time a 200-millicurie source was used, the samples being squares with areas increasing (from 0.71 to 2.4 cm per side) with the distance from the source. In this manner we obtained approximately equal initial activities at each point in the paraffin. The activity was kept sufficiently low that no correction for resolving time would be necessary. The statistical error was reduced to about one percent by repetition of the measurements.

The combination of the two series gave us the activities of both isomers as functions of the distance from the source, referred to a one-Curie source and a standard sample of surface $(2.4)^2$ cm².

For rhodium as well as for bromine, the distances in the paraffin were corrected for the sizes of the source and the sample in a manner similar to that of Rush.[12]

The irradiation times were practically infinite for the different half-lives, except for the 4.4-hr activity, which was corrected for infinite irradiation. The resulting counting-rates were measured by a Geiger-Müller counting system.

The initial counting rates, *viz.* the activities of both isomers at the end of the irradiation time, were determined from the total measured activity in the classical way. The counting times were long compared with the half-lives. In this way, the statistical errors were minimized,[13] because the deviation for a simple element is of the form:

$$\sigma = \{A_{1\,2} \exp[-\lambda(t_1 - t_2)]\}^{\frac{1}{2}},$$

where $A_{1\,2}$ is the total activity measured in the time from t_1 to t_2, so that the exponential is much smaller than unity if the time interval is large.

The determination of the mean square of the migration distance: $\langle r^2\rangle_{Av}$, was based on the classical formulas, fitted to each isomer:[2,12]

$$\langle r^2\rangle_{Av} = \left(\int_0^\infty \mathrm{Ar}^4 dr\right) \Big/ \left(\int_0^\infty \mathrm{Ar}^2 dr\right),$$

where A is the activity, and r is the distance from the source. These integrals are resolved into two parts. First, a graphical integration on the experimental points between 0 and 9 cm. For further distances the $\mathrm{Ar}^2(r)$ curve is considered to be exponential of the form e^{-ux}.

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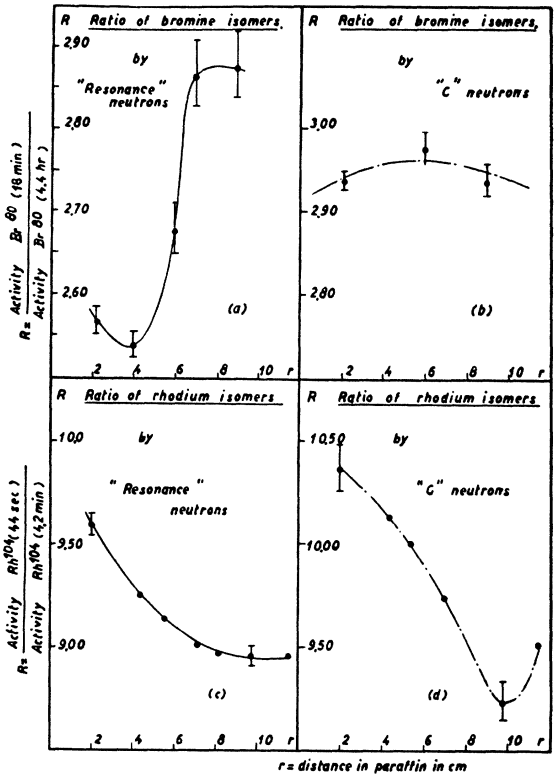

FIG. 1. Ratio of the saturated activities of the nuclear isomers of Br^{80} and Rh^{104} produced by resonance and by *C*-neutrons as a function of the distance from a Ra—Be neutron source in a paraffin medium. Distances are corrected for the sizes of the source and the sample. The indicated deviations are the probable statistical errors.

For bromine we used $u = 0.123$ cm^{-1} which is deduced from data by Rush.[12] For rhodium this coefficient was determined from our own experiments, where $u = 0.090$ cm^{-1} which is comparable to the data by Bakker.[14] This is why our values $\langle r^2\rangle_{Av}$ for Br and Rh are not directly comparable, but we are only interested here in the relative values for each isomer.

It would be interesting to find the real values of the resonance energy corresponding to each of the isomers, but this is not possible in our case, for the known relation;[2,15]

$$\ln(W/W') = [\langle r'^2\rangle_{Av} - \langle r^2\rangle_{Av}]/6\lambda\lambda'$$

will not give sufficient precision because of the importance of the possible errors in the absolute value of $\langle r^2\rangle_{Av}$, as well as the uncertainty in the values of the mean free paths λ, λ' corresponding to the resonance energies. Different authors give values fluctuating between 0.56 and 1.1 cm.[2,16–21] Probably the boron

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TABLE I. Saturated activities and ratio of activities of the isomers of Br^{80} and Rh^{104} produced by resonance and by *C*-neutrons, as a function of the distance from a Ra—Be neutron source in a paraffin medium.

Corrected distance	Resonance activity	Resonance activity	Ratio of resonance activities	Thermal activity	Thermal activity	Ratio of thermal activities
			Br(18 min)			Br(18 min)
r (cm)	Br(18 min)	Br(4.4 hr)	Br(4.4 hr)	Br(18 min)	Br(4.4 hr)	Br(4.4 hr)
2.22	395 ±4	153.5±0.7	2.57±0.02	1297 ±7	435 ±1	2.94±0.01
4.03	261 ±2	102.5±0.4	2.54±0.02			
5.97	144 ±2	53.7±0.4	2.68±0.03	790 ±6	265 ±1	2.98±0.02
6.95	112 ±1.8	38.9±0.3	2.87±0.04			
8.93	59.2±1.7	20.6±0.3	2.88±0.04	454.4±4	154.4±0.8	2.94±0.02
			Rh(44 sec)			Rh(44 sec)
r (cm)	Rh(44 sec)	Rh(4.2 min)	Rh(4.2 min)	Rh(44 sec)	Rh(4.2 min)	Rh(4.2 min)
2.04	1065 ±10	111 ±0.5	9.60±0.05	8480±128	817 ±6	10.38±0.12
4.36	677	73.2±0.4	9.24	7042	695 ±3	10.14
5.42	549	60.2	9.12	5747	575	10.00
7.11	322	35.7±0.3	9.02	4055	416.5±1.4	9.74
8.11	242.5	27	8.89	3321	348.5	9.53
9.77	150.5±2	16.8±0.2	8.96±0.05	2154±35	233.3±0.4	9.23±0.10
11.49	98.1±1.9	9.9±0.2	8.96±0.16	1411	148	9.53

method might give some results if applied with sufficient precision.[22]

III. RESULTS

The results for the initial counting rates and their ratios are given in Table I for bromine and for rhodium and are plotted in Fig. 1. Table II gives the cadmium ratio's for both elements at increasing distances.

Our conclusions from these results are as follows.

For *bromine* activated by the *resonance neutrons*, the ratio of the activities of the two isomers varies from 2.57±0.02 to 2.88±0.04 as the distance in the paraffin goes from 2 to 9 cm. As this ratio undoubtedly varies with the neutron energy, this seems to confirm the existence of more than one capture level. Furthermore, we can say that the lower capture level favors the formation of the ground state bromine isomer (18 min), in the energy region investigated, because the ratio increases with decreasing energy. These views are confirmed by finding two different values for the mean square migration distances, corresponding to each of the isomers:

$$\langle r^2\rangle_{Av}=97.8 \text{ cm}^2 \text{ for Br(18 min)},$$
$$\langle r^2\rangle_{Av}=94.4 \text{ cm}^2 \text{ for Br(4.4 hr)},$$

the first of which corresponds to the lowest resonance energy.

The activation of bromine by *C-neutrons* results in a ratio of activities which remains quite constant as the distance in the paraffin goes from 2 to 9 cm. This again is confirmed by the values of the mean square distances, which are quite the same for the two isomers.

However, a difference certainly exists between the relative activations of both isomers, on one side by thermal neutrons and on the other side by resonance neutrons. This result confirms a previous paper[6] and indicates that capture levels in the nucleus may be multiple.

In the case of *rhodium*, for the *resonance neutrons*, the ratio of activities of isomer Rh(44 sec) to Rh(4.2 min) varies from 9.60±0.05 to 8.96±0.05 at distances from the source from 2 to 10 cm. This indicates, as for bromine, but less precisely, the existence of more than one capture level, in agreement with Goldsmith and Rasetti,[7] who found by the boron method resonance energies of 1.16 ev for Rh(44 sec) and about 1 ev for Rh(4.2 min).

Here, in contrast to the case of bromine, the ratio of activities decreases with increasing distances from the source, so that it is the upper capture level which favors the formation of the ground-state isomer, Rh(44 sec), in the inspected region.

On the other hand, different authors[1,2,4,6] assume the existence of only one capture level, but they consider that the ratio of activities is the same with and without Cd. However, in these experiments only one distance in the paraffin appears to have been used. In our case too, at 10 cm the values of the "thermal" and the "resonance" ratio are nearly equal, but at other distances they differ.

For the corresponding $\langle r^2\rangle_{Av}$ values we find a very small difference, but come to the same conclusions as above.

TABLE II. The cadmium ratio C_R=activity without Cd/activity with Cd for the isomers of Br^{80} and Rh^{104}, as a function of the distance from a Ra—Be neutron source in a paraffin medium.

r (cm)	Cd ratio Br(18 min)	Cd ratio Br(4.4 hr)	r (cm)	Cd ratio Rh(44 sec)	Cd ratio Rh(4.2 min)
2.22	4.23	3.84	2.04	8.96	8.36
—	—	—	4.36	11.40	10.49
—	—	—	5.43	11.47	10.50
5.97	6.49	5.94	7.11	13.59	12.66
—	—	—	8.11	14.69	13.91
—	—	—	9.77	15.32	14.89
8.93	8.68	8.50	11.49	16.84	15.89

[22] (To be published later by the authors.)

The action of the *C-neutrons* on rhodium gives an unexpected result. Within the thermal region, the ratio of activities varies from 10.38 ± 0.12 to 9.23 ± 0.10 for paraffin-distances from 2 to 10 cm. This shows the existence of different capture levels in the *C*-neutron band, which seems to be due to resonances within the thermal region. Such resonances have already been proposed by others[7] and seem to be the more probable for Rh as this element has a resonance level at 1.33 ev which is close to the thermal region.

In this part of the experiments too, the ground state is favored by the higher energies, which is the same as observed in the resonance region.

The existence of different *C*-neutron capture levels seems to be confirmed by the different values obtained for the mean square distance of each isomer

$\langle r^2\rangle_{Av}=267.0$ cm^2 for the 4.2-min Rh isomer,
$\langle r^2\rangle_{Av}=263.1$ cm^2 for the 44-sec Rh isomer,

this value corresponding to the highest energy.

Finally we may notice that for fast neutrons (source without paraffin) the ratio of the isomer activities for bromine[6] was found to be 2.1 and for rhodium[5] about 6. The latter value was confirmed by us.

IV. CONCLUSIONS

If there is a single neutron capture level for nuclear isomers, this single level can but form both isomers in a invariable proportion.

As the ratio of the isomers varies with the distance from the source, there is evidence for the existence of *more than one* capture level. This is true for the resonance neutrons in bromine and perhaps in rhodium, whereas for rhodium, resonance levels seem to extend within the region defined by the *C*-neutrons. These views are confirmed by the fact that: (a) corresponding migration distances differ and (b) the ratio of the isomers certainly is different for resonance and for *C*-neutrons.

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An Experiment on the Anomalous Scattering of μ-Mesons by Nucleons

E. AMALDI AND G. FIDECARO
Istituto di Fisica della Università, Centro di Studio per la Fisica Nucleare del C.N.R., Rome, Italy
(Received August 7, 1950)

Large angle scattering of fast μ-mesons by an iron plate, 6 cm thick, has been investigated by means of a counter-hodoscope which recorded separately two energy bands: from 200 Mev to 320 Mev kinetic energy, and from 320 Mev to infinity. About half a million incident μ-mesons were counted. Upper limits for the cross sections for anomalous scattering were obtained: about 4.5×10^{-29} cm^2/nucleon for the low energy band; about 2.3×10^{-30} cm^2/nucleon for the high energy band. Also an upper limit for the cross section for production of penetrating showers by μ-mesons at sea level is given. We found a cross section of 10^{-30} cm^2/nucleon for showers of at least two particles of which at least one is emitted at an angle larger than 20° with a range of at least 7 cm Fe+5 cm Pb.

I. INTRODUCTION

IN a previous paper[1] we pointed out that the investigation of anomalous scattering of μ-mesons by nucleons seems unnecessary because of the well known experiment of Conversi, Pancini, and Piccioni, which showed that μ-mesons at rest interact very weakly with nucleons. However, it is not evident that the conclusions regarding the nuclear interactions between μ-mesons almost at rest and nucleons can be extrapolated to the case of μ-mesons with kinetic energy around several hundred Mev. In fact, Evans and George, and George and Trent,[2] working at sea level, under a clay thickness equivalent to 60 m water, found a local production of stars and penetrating showers which had to be due to μ-mesons. The corresponding cross section for these effects turned out to be of the order of 10^{-29} cm^2/nucleon; i.e., a value enormously larger than that expected according to the calculations of Fermi, Teller, and Weisskopf[3] on the capture of slow mesons by light nuclei. It is still smaller, however, than the lowest value yet obtained by means of direct observation of the anomalous scattering.[4] For these reasons we thought the anomalous scattering of mesons at sea level to be worthy of investigation. Some of the results reported here have already been published elsewhere.[1]

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