

AN INVESTIGATION OF (n, 2n) REACTIONS LEADING TO ISOMER FORMATION

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Data are presented on the cross sections for (n, 2n) reactions involving 14.7-Mev neutrons and resulting in the formation of the isomers Y^{88m} ($\sigma_m > 0.4$ barn), Nb^{92g} ($\sigma_g = 0.53 \pm 0.06$ barn), Pb^{207m} ($\sigma_m = 1.7 \pm 0.3$ barn), and Bi^{208m} ($\sigma_m = 0.66 \pm 0.12$ barn). The cross section for the reaction with lead was evaluated on the assumption that the isomer is produced only in the reaction $Pb^{208}(n, 2n)Pb^{207m}$. This value is compared with that computed from strong-interaction theory, assuming a two-stage mechanism of neutron evaporation in the (n, 2n) reaction. The Pb^{207} level scheme which agrees with the shell model was employed in the calculations. The relative probabilities of different types of transitions were estimated from the relation between the lifetime of an excited nuclear state and the transition energy. There is good agreement between the measurements and calculations. Radiation from the following isomers is reported: Nb^{92g} ($T_{1/2} = 10.0 \pm 0.3$ day, $E_\gamma = 0.94 \pm 0.01$ Mev), Bi^{208m} ($T_{1/2} = 2.6 \pm 0.1$ millisecc, $E_\gamma = 0.50 \pm 0.02$ Mev, $E_\gamma = 0.88 \pm 0.02$ Mev), and Na^{24m} ($T_{1/2} = 18.3 \pm 0.6$ millisecc).

PROCEDURE

OUR experimental technique and preliminary estimates of the cross sections for reactions with fast neutrons resulting in the formation of isomers with millisecond half-lives are similar to those described in reference 1. We have also published² an account of certain changes in technique together with the cross section obtained for the reaction producing Pb^{205m} . The technique described in reference 2 was used to measure the cross sections for the reactions producing Pb^{207m} and Bi^{208m} . The present paper gives the results of these measurements and data for additional isomers.

EXPERIMENTS

Samples with natural isotopic content were irradiated with neutrons produced in the pulsed bombardment of a thick tritium-zirconium target by D_2^+ ions from a 500-kv accelerating tube. The neutron spectrum was computed; the neutrons interacting with Pb, Nb, and Bi had a peak at $E_0 = 14.7$ Mev and a 0.25-Mev half-width.

γ rays from the samples induced pulses in a FÉU-S photomultiplier used in conjunction with a NaI(Tl) crystal; these pulses were then fed to amplitude and time analyzers during the intervals between neutron pulses. The cross section for a given reaction was determined by counting the

pulses forming the photopeak of total isomeric emission in the pulse-height distribution.

The efficiency of γ -ray registration by means of the photopeak pulse counts was determined with radioactive Cs^{137} , Nb^{95} , Nb^{92} , and Zn^{65} samples, whose activities were measured by two methods that produced mutually agreeing results. One method, as in reference 1, employed a counter with a special sheath, described by Sakharov;³ the other method used a NaI(Tl) crystal and spectrometer. In the latter case the efficiency of γ -ray registration was computed with the aid of data given in references 4 and 5. We estimated a 7 to 8% inaccuracy in determining the activities.

The yield of a given reaction as determined by the monitor was compared with the yield of $Cu^{63}(n, 2n)Cu^{62}$, for which the mean cross section 0.62 ± 0.04 barn was obtained from the computed neutron spectrum. The cross section for the investigated reaction was then calculated from the formula given in reference 1.

RESULTS

Our data give a half-life of 0.81 ± 0.02 sec for Pb^{207m} , in agreement with the review article of Strominger et al.⁶ The cross section for the reaction with lead was determined using a 1.04-Mev photopeak, for which pulses falling in 0.768 ± 0.006 sec intervals were discriminated. The start of these intervals was delayed for the time t (0.09

$\leq t \leq 0.11$ sec) after the leading edge of the ion pulse of the accelerating tube.

In interactions between 14-Mev neutrons and heavy nuclei a (n, 2n) reaction is the dominant inelastic process. For lead, bismuth, and certain other elements this has been confirmed by Ashby et al.,⁷ who reported a cross section for Pb (n, 2n) equal to the cross section for all inelastic processes: $\sigma_{(n,2n)}/\sigma_{(n,x)} = 1.07 \pm 0.08$. We assumed that in our experiments Pb^{207m} was formed only in a (n, 2n) reaction, and attributed our results to Pb^{208} . We obtained $\sigma_m^{\text{meas}} = 1.7 \pm 0.3$ barns for the $\text{Pb}^{208}(n, 2n)\text{Pb}^{207m}$ reaction, and compared this result with the calculated cross section σ_m^{calc} .

The cross section for $\text{Bi}^{209}(n, 2n)\text{Bi}^{208m}$ was determined similarly. γ rays with 0.88 ± 0.02 Mev and 0.50 ± 0.02 Mev, and a half-life of 2.6 ± 0.1 millisecond, were observed in the decay of Bi^{208m} . The pulse count gave a 0.88-Mev photopeak and cross section $\sigma_m = 0.66 \pm 0.12$ barn.

We also measured the cross section for the reaction producing a long-lived niobium isomer, and improved the half-life of Na^{24m} . The Nb^{92g} samples used to determine the efficiency of γ -ray registration were prepared by neutron bombardment of thin metallic niobium plates. We obtained 10.0 ± 0.3 days for the half-life of this isomer and detected 0.94 ± 0.1 Mev γ rays accompanying β decay. The experiments in which the reaction yield was determined from the radioactivity of the sample have already been discussed. We used the decay schemes of Nb^{92m} and Nb^{92g} given by Dzhel'epov and Peker.⁸ The cross section for $\text{Nb}^{93}(n, 2n)\text{Nb}^{92g}$ was $\sigma_g = 0.56 \pm 0.06$ barn.

Na^{24m} was formed in the reaction $\text{Al}^{27}(n, \alpha)\text{Na}^{24m}$. γ rays from the isomer were registered by a spectrometer used with a NaI(Tl) crystal whose packing contained neither aluminum nor magnesium. There was no background which could have been due to the reaction $\text{Na}^{23}(n, \gamma)\text{Na}^{24m}$. An improved value of 18.3 ± 0.6 msec was obtained for the half-life of Na^{24m} given in reference 1.

From earlier data¹ obtained with 14.9-Mev neutrons we estimated the cross section for the reaction producing the isomer with 14-millisecond half-life in the fast-neutron bombardment of yttrium. The result $\sigma_m > 0.4$ barn provided a sufficient basis for assuming that the given isomer resulted from a (n, 2n) reaction and for attributing the 0.24-Mev emission to Y^{88m} .¹

CALCULATION OF THE CROSS SECTION FOR $\text{Pb}^{208}(n, 2n)\text{Pb}^{207m}$

The cross section for the reaction producing Pb^{207m} was calculated on the basis of the strong-interaction theory and two-stage neutron evaporation. We calculated the partial cross sections for the formation of $\text{Pb}^{208} + n$ compound nuclei in states with spin j , as follows:

$$\sigma(j) = \pi\lambda^2(2j+1) \sum_{s=I_0-1/2}^{I_0+1/2} \sum_{l=|j-s|}^{j+s} T_l(E_0)/2(2I_0+1), \quad (1)$$

where $T_l(E)$ is the centrifugal barrier transmission coefficient for neutrons with orbital angular momentum l and energy E , $I_0 = 0$ is the spin of Pb^{208} , and $1/\lambda = k$ is the wave number of a neutron with energy E_0 . We then calculated the partial cross sections for the formation of Pb^{208*} in states with spin j_1 following the evaporation of the first neutron:

$$\sigma(j_1) = \sum_j \sigma(j) \left[\sum_{s_1=j_1-1/2}^{j_1+1/2} \sum_{l_1=|j-s_1|}^{j+s_1} \int_0^{E'_{\text{max}}} T_{l_1}(E') f(E') dE' \right] \times \left[\sum_{l_q} \sum_{s_q=j_q-1/2}^{j_q+1/2} \sum_{l_q=|j-s_q|}^{j+s_q} \int_0^{E'_{\text{max}}} T_{l_q}(E') f(E') dE' \right]^{-1}, \quad (2)$$

where $f(E') dE' = \text{const} \times E' \exp(-E'/\tau_1) dE'$ is the energy distribution of neutrons evaporating in the first stage, at the temperature $\tau_1 = 1$ Mev.

Equations (1) and (2) were derived from the equation of Hauser and Feshbach⁹ that gives the cross section for the excitation of an individual level in inelastic neutron scattering. We separated the parts pertaining to the formation and decay of the compound nucleus, and integrated over the energies of evaporating neutrons instead of summing over the energies of separate levels. It was assumed that compound nuclei decay into close energy levels. In Eq. (2) the spin dependence of level density was taken into account by the summations over $l_{q(1)}$ and $s_{q(1)}$, and the dependence on excitation energy was taken into account by the function $f(E')$.

We kept in mind the remark of Remy and Winter¹⁰ that τ_1 must be 20–30% above the temperature determined experimentally (0.7–0.8 Mev according to references 11 and 12), neglecting two-stage evaporation. We also assumed that τ_1 can differ little from the result 1.05 Mev obtained by Rosen and Stewart for the Bi (n, 2n) reaction.¹³

It can be assumed that with the evaporation of a second neutron Pb^{208*} compound nuclei decay to states of Pb^{207*} , where the level density is suffi-

cient to permit the use of a near-Maxwellian energy distribution in the calculations. Neutrons in the range 5–12 Mev were not observed in the case of the reactions with lead reported in references 11 and 12. These neutron energies would have been observed if decay to discrete levels were dominant. There are both small and large spin values among the low-energy levels of Pb^{207} . The probability of decay to these levels, which contains the factor $T_l(E')$, would be relatively large.

Keeping the foregoing considerations in mind, we used (2) to calculate the spin distribution of Pb^{207*} nuclei formed after the evaporation of a second neutron. j_1 and j were replaced by I^* and j_1 , respectively, and the energy distribution was characterized by the temperature $\tau_2 = 0.5$ Mev, which is close to the values 0.4_6 and 0.5_3 Mev estimated by Rosen and Stewart¹³ for the Ta ($n, 2n$) and Bi ($n, 2n$) reactions.

Penetration of the centrifugal barrier was calculated from

$$[T_l(x)]^{-1} = \frac{1}{2} + \frac{\pi}{8X} \left\{ (X^2 + l^2) [J_{-(l+1/2)}^2(x) + J_{l+1/2}^2(x)] + x^2 [J_{-(l-1/2)}^2(x) + J_{l-1/2}^2(x)] + 2lx [J_{-(l+1/2)}(x)J_{-(l-1/2)}(x) - J_{l+1/2}(x)J_{l-1/2}(x)] \right\}, \quad (3)$$

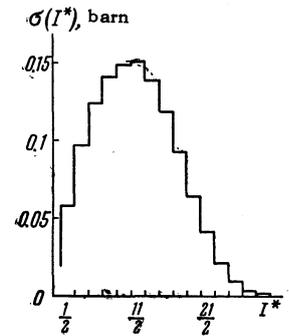
where $x = kR = 0.222\sqrt{E(\text{Mev})} \times 1.45 A^{1/3}$, $X^2 = x^2 + X_0^2$, and $J_{\pm(l \pm 1/2)}(x)$ is the Bessel function of the first kind and of order $\pm(l \pm 1/2)$. X_0^2 is expressed in terms of the depth V_0 and radius R of the rectangular potential well representing the potential in the simplest type of strong interaction; thus $X_0^2 = 2mV_0R^2/\hbar^2$. Equation (3) was obtained from the book by Blatt and Weisskopf,¹⁴ with $v_l(x)$ and $v_l'(x)$ replaced by Bessel functions. We used the value $X_0^2 = 144$, with which Oleksa¹⁵ obtained best agreement between the calculated cross section for $\text{Pb}^{207}(n, n')\text{Pb}^{207m}$ and experimental data on inelastic neutron scattering at $1.6 \leq E_0 \leq 2.5$ Mev. It was estimated that with $X_0^2 = 64$, $\sigma(I^*)/\sum_{I^*} \sigma(I^*)$ can exceed the result obtained with $X_0^2 = 144$ by no more than 5%. The results obtained for $\sigma(I^*)$ are shown in the figure. Identical partial cross sections are obtained for the formation of Pb^{207*} in states with different parities.

In view of the results obtained by Ashby et al.⁷ we used

$$\sigma_{(n, 2n)}^{\text{calc}} = \sum_j \sigma(j) = \sum_{I^*} \sigma(I^*), \quad \sigma_m^{\text{calc}} = \sum_{I^*} r_m(I^*) \sigma(I^*)$$

to represent the cross sections for $\text{Pb}^{208}(n, 2n)\text{Pb}^{207*}$ and $\text{Pb}^{208}(n, 2n)\text{Pb}^{207m}$, respectively. $r_m(I^*)$ and $r_g(I^*)$ are the respective probabilities that a nucleus in a highly excited state with spin I^* drops to a metastable state ($13/2^+$) or

Partial cross sections for the formation of Pb^{207*} nuclei in states with the same parity.



directly to the ground state [$r_m(I^*) + r_g(I^*) = 1$]. These probability coefficients were evaluated from Montalbetti's nomogram¹⁶ relating the mean lifetime of an excited nuclear state with the type and energy of a single-particle transition accompanied by γ -ray emission. It was assumed that this nomogram is applicable to the relative probabilities of different transition types independently of the nuclear model. It was also considered that from states with $E^* \approx 6.5 - 7$ Mev, including possible non-single-particle states, transitions proceed to single-particle Pb^{207} levels determined from the study of radioactive decay, given by Dzhelepov and Peker,⁸ and proposed by Harvey¹⁷ from an investigation of (d, p) and (d, t) reactions. The arrangement of these levels agrees with the shell model, as follows: $p_{1/2}$ (ground state); $f_{5/2}$, 0.570 Mev; $p_{3/2}$, 0.894 Mev; $i_{13/2}$, 1.633 Mev (metastable state); $f_{7/2}$, 2.34 Mev; $g_{9/2}$, 2.75 Mev; $i_{11/2}$, 3.60 Mev; $d_{5/2}$, 4.42 Mev; $g_{7/2}$, 4.66 Mev; $d_{3/2}$, 5.28 Mev. We allowed the existence of a $15/2^-$ level at ~ 5 Mev, assumed to result from the strong splitting of a level with orbital moment 7 due to spin-orbit interaction.

The values obtained for $r_m(I^*)$ by the described procedure are as follows. For all even states with spins $I^* \geq 11/2$ and odd states with spins $I^* \geq 7/2$, $r_m(I^*) = 1$. For all other states forming the distribution shown in the figure, $r_m(I^*) = 0$. It was here assumed that the probabilities of different types ($E1, M1, E2, \dots$) of transitions were comparable if they did not differ by a factor greater than 20. For $E^* \gtrsim 6$ Mev there was no competition between transitions of the different types, and $r_m(I^*)$ was independent of E^* .

It was to be expected that the coefficients $r_m(I^*)$ would be close to unity or zero and independent of the excitation energy. Katz and others¹⁸⁻²⁰ investigated the relative yields σ_g/σ_m for several isomers formed in (γ, n) reactions and found σ_g/σ_m to be practically constant over broad intervals of γ -ray energy. Dipole absorption predominated; it could therefore be assumed that the spin distributions of excited nuclei were only slightly dependent on energy, and the constant

value of σ_g/σ_m could be accounted for by the fact that $r_{m(g)}(I^*)$ is independent of E^* . The summary of data in reference 21 gives the relative yields of isomers resulting from thermal neutron absorption by even-even nuclei; in the great majority of cases $r_{m(g)}(1/2^+)$ is close to unity. $r_{m(g)}(I^*) \approx 1$ from the results of experiments in which isomers were formed in (n, γ) reactions with thermal neutrons and with fission neutrons.²²⁻²⁴

The given values of $r_{m(I^*)}$ yielded $\sigma_m^{\text{calc}} = 1.58$ barns, which is in satisfactory agreement with σ_m^{meas} . We also obtained $(\sigma_m/\sigma_{n,2n})_{\text{calc}} = 0.65$; considering that this is only slightly dependent on X_0^2 , we calculated the cross section $\sigma_{(n,2n)}^0 = \sigma_m^{\text{meas}}(\sigma_{(n,2n)}/\sigma_m)_{\text{calc}} = 2.6$ barns for Pb^{208} (n, 2n) Pb^{207*} . Within the limits of experimental and calculational accuracy this result agrees with $\sigma_{(n,2n)} = 2.74 \pm 0.20$ barns in reference 7, and with the cross sections for inelastic interactions between neutrons with 14.2–14.5 Mev and lead nuclei, for which the results $\sigma_{(n,x)} = 2.56 \pm 0.03$ barns and 2.54 ± 0.05 barns are given in reference 25 and 26, respectively.

Natural lead was used in references 7, 25, and 26. Our results therefore permit application of the strong-interaction model and the roughly approximate statistical theory to the (n, 2n) reaction in the case of the double magic isotope Pb^{208} . The agreement of $\sigma_{(n,2n)}^0$ with $\sigma_{(n,2n)}$ and $\sigma_{(n,x)}$, and of σ_m^{calc} with σ_m^{meas} , show that in analyzing the spin distributions of excited nuclei formed in reactions it is sometimes permissible to use the aforesaid procedure for calculating the coefficients $r_{m(I^*)}$.

It has been shown in references 27 and 28 that the correlations of proton and neutron positions in nuclei can result in a very low probability of E1 transitions in the case of low excitation energies. This evidently applies to Pb^{207} , where the $g_{9/2} \rightarrow i_{13/2}$ transition, for example, has considerably higher probability than the $g_{9/2} \rightarrow f_{7/2}$ transition. We therefore repeated our calculation excluding electric dipole transitions in evaluating $r_{m(I^*)}$. We obtained $r_{m(I^*)} = 1$ for all odd states with spins $I^* \geq 13/2$ and even states with $I^* \geq 11/2$. For $I^* = 3/2^+$ and $7/2^+$ competition is possible between M1 transitions (with relatively small transition energies) and M2 transitions (with transition energies ~ 6 Mev). For $I^* = 3/2^+$ and $7/2^+$ we have $r_{m(I^*)} = 0.2$. For $I^* = 9/2^+$ the M1, E2, and M2 transition probabilities are comparable and $r_{m(9/2^+)} = 0.9$. For the other states represented in the spin distribution shown in the figure we have $r_{m(I^*)} = 0$. The final results are $\sigma_m^{\text{calc}} = 1.32$ barns and $\sigma_{(n,2n)}^0 = 3.1$ barns. These val-

ues agree with the values of σ_m^{meas} , $\sigma_{(n,2n)}$, and $\sigma_{(n,x)}$ given above. However, since the agreement was worsened when E1 transitions were excluded we consider it highly probable that when $E^* \gtrsim 3.6$ Mev in Pb^{207} , electric dipole transitions play an important part.

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