Neutron-induced gamma-ray production in ²⁰⁸Pb[†]

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The cross section for γ -ray emission associated with inelastic scattering of neutrons in the 208 Pb $(n, n'\gamma)$ 208 Pb reaction has been measured at several angles for neutron energies between 3.1 and 5.25 MeV. The experimental data have been compared with calculations based on the Satchler formalism, with allowance for width-fluctuation corrections. Agreement for the 2.61-MeV γ ray is excellent throughout. The experimentally observed cross sections for the other γ rays were generally 30 to 50% below the theoretical values, except for the 0.583-MeV γ ray, for which the observed values were well above the theoretical values. Resolution of several γ -ray peaks with a Ge(Li) detector has led to a modified level scheme for ²⁰⁸Pb.

NUCLEAR REACTIONS ²⁰⁸Pb $(n, n'\gamma)^{208}$ Pb, E = 3.1 - 5.25 MeV, measured $\sigma(E, \theta)$ for deexcitation γ rays. Compound nuclear theory analysis, including width fluctuations.

I. INTRODUCTION

The ²⁰⁸Pb nucleus with closed shells of both neutrons and protons and the only doubly magic nucleus in the heavy-element region of the Periodic Table is of considerable experimental and theoretical interest. A summary¹ of the extensive data is presented in various compilations, and no attempt will be made to review here the complex situation regarding this nucleus.

In the present study we have sought to obtain more information on the location and characteristics of excited levels of ²⁰⁸Pb which lie below 5 MeV. Toward this end, γ -ray production cross sections and γ -ray angular distributions from neutron inelastic scattering were measured using incident neutron energies lying between 3.1 and 5.25 MeV. The origins of the prominent γ -ray peaks. observed are discussed in light of the known level scheme, and the experimentally determined cross sections and angular distributions of the more prominent γ rays are compared with values obtained from theory.²⁻⁴

II. EXPERIMENTAL PROCEDURE

Monoenergetic neutrons were produced by the 2 H(d, n)³He reaction in the Texas Nuclear 3.2-MeV Van de Graaff accelerator. The target consisted of a 1-cm-long gas cell separated from the vacuum system by a 2.59-mg/cm² molybdenum foil which was filled with deuterium to a pressure of 1 atm

for the lower-energy measurements and 2 atm for the higher measurements. The neutrons were scattered from cylindrical scattering samples aligned with their longitudinal axes at right angles to the direction of the neutron beam. Their centers were positioned about 5-7 cm from the center of the target. The energy spread of the incident neutrons comes principally from the thickness of the gas target and from the energy variation associated with the angular spread of the neutrons over the scatterer. A small additional spread came from energy straggling in the target entrance foil. The average spread in the neutron energy consisted of ±100 keV due to target thickness, ±50 keV from the angular spread of neutrons over the scatterer, and ±20 keV due to straggling in the target entrance foil. Scattering samples consisted of isotopically enriched (99.75%) cylinders of ²⁰⁸Pb on loan from Oak Ridge National Laboratory. Three different samples, having diameters between 1.6 and 2 cm and lengths between 2 and 4.5 cm, were used in the course of the experiments.

Scattering samples were placed at 0° with respect to the direction of the incident chargedparticle beam and the γ -ray spectrometer was rotated about the vertical axis of the scattering samples. The sample to detector distance varied between 100 and 120 cm. A tungsten and iron shadow bar prevented direct neutrons from the target cell from reaching the detector. The neutron flux was monitored at either 0 or 90° to the incident beam by a calibrated long counter whose absolute

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efficiency had been established by comparison with a proton-recoil telescope. An auxiliary monitor consisting of a biased plastic scintillator was placed at 45° to the incident beam for some of the measurements. Additional measurements were made at the end of each data run to determine the factors needed to correct the long-counter results for the effects of room background and scattering from the ²⁰⁸Pb samples.

The experimental detection equipment, which has been described elsehwere,^{5, 6} consists basically of a two-crystal anticoincidence spectrometer operated in conjunction with conventional neutron timeof-flight techniques.⁷ The anticoincidence operation of the spectrometer considerably reduces the Compton-scattering component of the photons interacting with the main detector, a feature which greatly facilitates the interpretation of complex spectra. Neutron background effects are significantly suppressed by both the time-of-flight and anticoincidence gating methods employed. Most of the measurements were made using this system, but for a few measurements the center NaI(Tl) detector was replaced with a 34-cm³ solid-state Ge(Li) detector which became available near the termination of this experiment.

The γ -ray spectra produced by $(n, n'\gamma)$ interactions in the scatterer were obtained by collecting data in a multichannel analyzer (MCA) only during a suitably selected interval following the bombarding beam pulse. These are called the "foreground spectra." A second MCA accumulated "background spectra" at time intervals which were adjusted to exclude the detection of γ rays which were directly associated with the inelastic scattering by the scattering sample.

III. ANALYSIS

The foreground spectra obtained for a particular value of E_n were corrected by subtracting the corresponding background spectra. The γ -ray yield for each of the prominent peaks in the difference spectrum was determined by integration of the spectrum over the region of interest and applying additional corrections. These included corrections for self-absorption of the γ rays by the ²⁰⁸Pb scatterer, for detector efficiency, and for Comptonscattering contributions from higher-energy γ rays. The effective neutron flux was determined by correcting the value obtained from the long counter for anisotropy over the sample and for neutron attenuation in the sample. Multiple-scattering estimates, based on a second-scattering event of nonelastic character by neutrons that have elastically scattered once, indicate that the effective neutron flux is increased by less than 0.5%. Both

attenuation and multiple-scattering calculations were made using the technique of Cranberg and Levin.⁸

Theoretical comparisons to the data were made on the basis of compound nuclear (CN) theory using the formalism developed by Satchler.^{3, 4} Transmission coefficients which included spin-orbit effects were first calculated from the computer code ABACUS,⁹ using the potential parameters of Rosen.¹⁰ These transmission coefficients were then used in the computer code TRNRX¹¹ to calculate cross sections and angular distributions. Calculations were made both with and without width fluctuations using the computer code NEARREX of Moldauer.¹²

In order to make any of the calculations it is necessary to have (or evolve) a suitable energy-level diagram including the spin and parity of each level. In the case of ²⁰⁸Pb the characteristics of the ground state and the first four excited states are well established. These states, as well as the next five additional excited states in the region above



FIG. 1. Energy-level and deexcitation scheme for 208 Pb based on correlation between previous work and our results. The 1.44- and the 4.08-MeV γ rays have not been previously observed. Transition energies in some cases do not correspond exactly to level differences; the absolute discrepancies are, however, within our errors of measurement.

3.709 MeV are shown in Fig. 1. We have drawn on Ref. 1, including specifically certain related experimental studies, $^{13-17}$ in determining the location and characteristics of these states. The characteristics of the upper five states are listed in Ref. 1 as (4^-) , 2⁺, $(5)^-$, (2^-) , and $(4^-, 5^-)$, where the parentheses indicate uncertainty. Analysis of our data has shown consistency with all of these assignments, except that we originally chose to assign 3⁻ to the 4.231-MeV state. We have not had opportunity to repeat these calculations with a 2⁻ assignment.

The branching ratio of the 3.475 state has been reported by Earle *et al.*¹⁶; the other branching ratios (all 0.5/0.5) are based on the experimental data which we obtained at $E_n = 5.0$ MeV and which will be presented in the next section of this paper.

IV. RESULTS AND DISCUSSION

 γ -ray spectra were taken at 55 or 90° at six different neutron energies in the interval 3.1 MeV $\leq E_n \leq 5.25$ MeV. Observations were also made at several angles for each specific neutron energy.

A. Cross sections

The cross sections which were measured experimentally are shown in Table I; the corresponding theoretical values are also shown. These theoretical values include the width-fluctuation corrections. Without these, the theoretical values were 30 to 40% higher than those shown in the table. It has also been observed by Mathur, Buchanan, and Morgan¹⁸ that the inclusion of width-fluctuation effects reduces calculated cross sections by as much as 45%.

At the lowest values of E_n which are shown in Table I, viz., 3.1 and 3.5 MeV, only the first one or two excited levels of ²⁰⁸Pb are involved. As previously pointed out, the characteristics of these levels are well known, and the theoretical and experimental results which we obtained are readily compared since there are few decay channels open. Only the 2.615- and the 0.583-MeV γ rays are observed at these bombarding energies. Our experimental uncertainties consist of the statistical error in counts, a 7% uncertainty in determining the true photon yield, and a 7.5% uncertainty in determining the effective neutron flux. We therefore conclude that the agreement between theoretical and experimental results is quite satisfactory at these bombarding energies.

At neutron bombarding energies of 4.1 MeV and above the γ -ray spectra which result from inelastic scattering from ²⁰⁸Pb become much more complex. When only the NaI(Tl) detector was available, our attempts to fit experimental observations to assumed energy-level diagrams were generally disappointing. However, when the Ge(Li) detector became available, spectra were obtained with much higher resolution. In particular, the peaks in Fig. 2 at 0.76, 0.86, 1.37, and 1.63 MeV are seen to be doublets in Fig. 3. Another run made at 4.5 MeV gave a spectrum similar to that shown in Fig. 3 except that the doublet near 1.6 MeV was not observed.

Examination of Table I for measurements at $E_n \ge 4.1$ MeV reveals that the experimentally observed

$E_{\rm v}$ (MeV)) θ 2.615		0.583		0.86			0.749			0,763			
E'_n (MeV)	(deg)	Th.	Exp.	Th.	Exp.	Th.	E	xp.	Th.	Exp).	Exp).	Th.
3.1 ± 0.15	90	42.6	40 ± 4											
3.5 ± 0.2	90	61.5	52 ± 6	9.2	11 ± 2									
4.1 ± 0.15	90	90.0	90± 9	28.6	47 ± 5	13.9	12	±1.5	(a)	$[1.8 \pm 0.1]$		0.3]		3.0
4.5 ± 0.1	90	103.5	99 ± 10	33.5	63 ± 7	20.0	12 :	±1.4 4.7			[7.5±	.5±0.9]		5.8
5.0 ± 0.08^{b}	55	128.5	135 ± 14	45.1	58 ± 7	21.2	11	±1.3	6.6	$4.2 \pm$	0.5	3.9±	0.4	6.4
5.25 ± 0.08	90	113.0	121 ± 13	38.8	76 ± 8	23.8	11 :	±1.1 7.3		[9.8±	9.8 ± 1.1]		7.0	
E_{γ} (MeV)	θ	0.984 1.		.38 1.44		4	1.617			1.643		4.08		
E'_n (MeV)	(deg)	Th. Exp	. Th.	Exp.	Exp.	Th.	Th.	Exp.		Exp.	Th.	Th.	E	xp.
4.1 ± 0.15	90		3.3	[5.5	±0.8]	4.1								
4.5 ± 0.1	90		6.2	[7.5	±0.9]	9.5	3.7	[6	.7±0	. 9]	1.9			
5.0 ± 0.08^{b}	55	5.76 3.0±	0.6 6.4	3.9 ± 0.5	6.6 ± 1.0	10.2	6.6	4.3 ± 0.5	57	.8±0.8	5.4	10.3	6.6	±1.1
5.25±0.08	90		7.3	[7.7	± 0.9]	10.4	6.1	[12	.3 ± 1	.4]	4.9	9.1	6.4	±1.0

TABLE I. Differential cross sections (in mb/sr) for γ rays of various energies emitted in the ²⁰⁸Pb($n,n'\gamma$)²⁰⁸Pb reaction at various bombarding energies. The theoretical values include width-fluctuation corrections, as explained in the text. Brackets around entries indicate that it was not possible to resolve the experimentally observed lines; hence the value of such entries corresponds to the sum of the two associated entries in the theoretical columns.

^a Not excited at this bombarding energy.

^b Ge(Li) detector.



FIG. 2. γ -ray spectrum from ²⁰⁸ Pb at E_n = 5.25 MeV observed with a NaI(Tl) detector.

cross sections for emission of the 2.615-MeV γ ray are in full agreement with the theoretical calculations, within experimental error of about 10%. Good agreement is also obtained for the doublet at 1.617 and 1.643 MeV. The other experimental cross sections are generally 30 to 50% below the calculated values for $E_n \ge 4.1$ MeV, except for the 0.583-MeV γ ray. The experimental and theoretical cross sections for the 0.583-MeV γ ray agree only at the lowest bombarding energy. It might be supposed that at bombarding energies of 4.1 MeV and above, a second mode of deexcitation is contributing to the experimentally observed cross section. However, measurement of the γ -ray spectrum with the Ge(Li) detector (see Fig. 3) revealed that the 0.583-MeV peak was due to the deexcitation of a single level or to a doublet whose separation was less than the resolution of the detection system (\approx 11 keV).

Although the fourth excited state at 3.709 MeV decays by emission of a 0.511-MeV γ ray, the prominent γ ray we observed at this energy comes principally from other sources. Measurements made with scattering samples of bismuth and of ²⁰⁸Pb confirm this. Although bismuth has no deexcitation γ ray of this energy, prominent peaks having approximately the same amplitude at 0.511 MeV appear in both spectra. It is clear that some of the 0.511-MeV line is produced by background effects almost certainly associated with annihilation radiation. Attempts to explain its existence by assuming that pairs were produced by γ rays from the ²⁰⁸Pb deexcitation failed because the calculated intensities were much too low. For these reasons, no cross-section data for the 0.511-MeV γ ray are presented in Table I.



FIG. 3. γ -ray spectrum from ²⁰⁸Pb observed with a Ge(Li) detector. The small (0.849) peak is an iron background peak from the detector shadow bar.

B. Angular distributions

The angular distribution of the 2.61-MeV γ ray was obtained for neutron bombarding energies of 3.1, 3.5, 4.1, 4.5, and 5.25 MeV. The most complete data were obtained at $E_n = 4.1$ MeV; these are shown in Fig. 4. Data were taken with both the NaI(Tl) and the Ge(Li) detector with close agreement between these observations. The agreement between theory and experiment here is very good, as it was at all of the other bombarding energies, except at the lowest. For this case ($E_n = 3.1$ MeV) the observed cross sections were lower than calculated values by approximately 25% at 30°, the smallest angle of observation.

The observed angular-distribution data for the 0.583-MeV γ ray showed satisfactory agreement with theory at E_n values of 3.5 and 4.1 MeV (see Fig. 5), although the statistical errors here were much greater than for the data on the 2.61-MeV γ ray. Angular-distribution data on the 0.583-MeV γ ray at a neutron bombarding energy of 4.5 and 5.25 MeV showed differential cross-section ratios $[W(\theta)/W(90^\circ)]$ which varied by only 10% or so, although the theoretical ratios at 0° should be nearly 50% above the value at 90°. It will be recalled that the 0.583-MeV γ ray also showed considerable variation between experimental and theoretical cross sections at 90° (Table I).

The angular-distribution data for the 0.86-MeV γ ray at $E_n = 4.5$ MeV are shown in Fig. 6. Theoretical distributions are shown in that figure for both M1 and E2 transitions. The data are consistent with a pure M1 transition. Data at $E_n = 4.1$ and 5.25 MeV agree with this conclusion.

Attempts to obtain satisfactory angular distributions for the 0.76- and the 4.08-MeV γ rays were thwarted by poor statistics.



FIG. 5. Angular distribution of the 0.583-MeV γ -ray peak at 4.1-MeV bombarding energy. The solid line is the theoretical curve for a pure M1 transition.

C. Prior data and our observations

The γ rays at 0.749, 0.984, 1.38, and 1.643 MeV have previously been reported by Pakkanen et al.¹⁷ in the decay of ²⁰⁸Tl. The 0.984- and 1.38-MeV γ rays were assigned the same transitions as those shown in Fig. 1; the other two γ rays were not given an assignment by these authors. A recent ²⁰⁷Pb- $(d, p\gamma)$ study by Earle *et al.*¹⁶ reported γ rays of 1.615 and 1.638 MeV from energy levels similar to those shown in Fig. 1. To our knowledge, the 1.44- and 4.08-MeV γ rays have not been previously reported. The two γ -ray transitions at 1.643 and 1.617 MeV in Fig. 1 probably correspond to those reported by Earle et al.¹⁶ since they appear to come from levels having nearly the same energies. The assignment of the 0.749- and the 1.44-MeV transitions was made on the basis of the level scheme of Fig. 1, although no branching has been reported¹⁶ from the level at 4.23 MeV.



FIG. 4. Angular distribution of the 2.61-MeV γ ray at $E_n = 4.1$ MeV. The solid line represents a theoretical calculation for a pure E3 transition. Triangles and circles refer to separate sets of measurements.



FIG. 6. Angular distribution of the 0.860-MeV γ ray at $E_n = 4.5$ MeV. Theoretical calculations are shown for pure **M1** and pure **E2** transitions.

V. CONCLUSIONS

The bombardment of ²⁰⁸Pb by neutrons of energy up to 4 MeV excites primarily only the levels at 2.615 and 3.198 MeV. In this region of bombarding energies, the measured cross sections and angular distributions of these deexcitation γ rays agree very well with theoretical calculations which are presented in this paper, provided width-fluctuation corrections are included.

At bombarding energies in the region 4.1 MeV $\leq E_n \leq 5.25$ MeV, the measured differential cross sections for deexcitation of the 2.615-MeV γ ray are in full agreement with theoretical values within the experimental error of about 10%.

At bombarding energies of 4.1 MeV and above the γ -ray spectra become increasingly complex, and the measured cross sections for deexcitation are generally below the calculated values, except

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for the 0.583-MeV γ ray. If this radiation comes from a doublet our data show that its members must be separated by $\simeq 11$ keV or less. The angular distribution of the 0.583-MeV γ ray is also in disagreement with theoretical values at $E_n \ge 4.5$ MeV.

The angular-distribution data for the 0.86-MeV γ ray are consistent with a pure M1 transition at the bombarding energies at which it is observed.

New deexcitation γ rays of energies 1.44 and 4.08 MeV were observed in the course of this work. Through high-resolution spectroscopy, it was found that the peaks in a NaI(Tl) spectrum at 0.76, 0.86, 1.38, and 1.63 MeV were actually doublets which are separated by approximately 14 to 60 keV. It is found that our experimental data may be reasonably fitted into a level scheme which we evolved and which is based on levels hitherto observed in various other reactions involving ²⁰⁸Pb.

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