Total Neutron Cross Section of Pb²⁰⁸

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Measurements are reported on the total neutron cross section of isotopically enriched Pb²⁰⁸ (99.75%) for neutron energies in the range from 720 to 1890 keV with an energy spread of about 3 keV. In this region at least 85 resonances are observed, of which 24 are analyzed to give tentative spin assignments and reduced widths. The reduced-width estimates furnish evidence that the J = 5/2 resonances at 723 and 821 keV have even parity. Differential cross sections of normal lead measured with 50-keV energy spread at 1.2, 2.2, and 3.2 MeV were used in estimating the in-scattering correction for the total cross-section data and are included in this paper.

INTRODUCTION

HE independent-particle model is expected to be applicable for much of the description of the interaction of a nucleon with a doubly-closed shell nucleus. Here the system, consisting of a tightly bound spherical core plus (or minus) one nucleon, gives rise to low-lying states for which the single-particle character is predominant. For the light doubly closed shell nuclei, most of the low-lying nuclear states in He⁵ and O¹⁷ and low-energy phase shifts for scattering of neutrons from He⁴ and O¹⁶ can be explained in terms of the interaction of the neutron with average phenomenological potentials.^{1–3} It is of considerable interest then to investigate the heaviest doubly closed shell nucleus, Pb²⁰⁸, from this point of view. With natural lead samples $\lceil Pb^{208} \rceil$ (51.4%), Pb²⁰⁶ (26.7%), Pb²⁰⁷ (20.6%), and Pb²⁰⁴ (1.3%) only a few neutron resonances assigned to Pb²⁰⁸ have been reported in the literature.^{4,5} By use of a radiogenic lead sample [Pb²⁰⁶ (88.3%), Pb²⁰⁷ (8.6%), and Pb²⁰⁸ (3.1%)] to compensate for the effect of the Pb²⁰⁶ in natural lead, some neutron scattering information has been accumulated on a mixture of isotopes of an effective composition: Pb²⁰⁸ (72%-70%), Pb²⁰⁷ (26%-28%), and Pb²⁰⁴ (2%).⁶⁻⁸ The isotopic composition [Pb²⁰⁸ (71.3%), Pb²⁰⁶ (26.7%), and Pb²⁰⁷ (2.0%)] of lead extracted from Tennessee shale makes it possible to use the compensation technique with radiogenic lead to acquire information on a sample of effective composition 99% Pb^{208,3,9} An investigation of the total cross section of Pb²⁰⁸ by use of this sample and with ~ 10 -keV resolution over the range 0.9 to 2.6 MeV, and with

 \sim 20-keV resolution up to 4.32 MeV has revealed much unresolved structure.³ These data make it apparent that better energy resolution is required to unravel the resonances for Pb208 neutron scattering. The recent availability of enriched Pb²⁰⁸ from the Isotopes Division of the Oak Ridge National Laboratory makes such relatively high resolution measurements feasible.

EXPERIMENT

The total cross section of separated Pb²⁰⁸ (99.75%)¹⁰ was determined by the standard transmission technique.¹¹ The $Li^7(p,n)Be^7$ reaction produced by protons from the 5.5-MV ORNL electrostatic accelerator served as the neutron source. Lithium metal was evaporated on a rotating tantalum surface; the target thickness was obtained by measuring with a long counter the rise in neutron yield at threshold.¹² The neutron energy scale was based on the $\text{Li}^7(p,n)\text{Be}^7$ threshold¹² and on the calibration of the proton beam analyzing magnet.¹³ The reproducibility of the energy position of the resonances shown in Fig. 1, together with previous experience with the 5.5-MeV proton beam, indicates that the energy scale of Fig. 1 is good to within ± 5 keV.

Neutrons were detected with 1-in.-diam stilbene crvstals from which pulses due to gamma rays were depressed by means of pulse-shape discrimination. For much of the data presented (particularly for that taken at the lower energies) the Brooks-Owen technique for pulse-shape discrimination was used.14 For the rest of the data the Forte circuit was used in conjunction with the stilbene crystal for this purpose.¹⁵ The center of the

¹ E. Van der Spuy, Nuclear Phys. 1, 381 (1956). ² J. L. Fowler and H. O. Cohn, Phys. Rev. 109, 89 (1958). ³ J. L. Fowler, E. G. Corman, and E. C. Campbell, p. 474, Proceedings of the International Conference on Nuclear Structure (University of Toronto Press, Toronto, 1960).

⁴ H. H. Barschall, C. K. Bockelman, R. E. Peterson, and R. K. Adair, Phys. Rev. **76**, 1146 (1949).

⁶ H. W. Newson, J. H. Gibbons, H. Marshak, R. M. Williamson, R. A. Mobley, A. L. Toller, and R. C. Block, Phys. Rev. **102**, 1580 (1956). ⁶ R. E. Peterson, R. K. Adair, and H. H. Barschall, Phys. Rev. **70**, 035 (1050).

<sup>79, 935 (1950).
7</sup> J. L. Fowler, Bull. Am. Phys. Soc. 5, 443 (1960).
⁸ R. M. Wilenzick, G. E. Mitchell, K. K. Seth, and H. W. Lewis, Phys. Rev. 121, 1150 (1961).
⁹ E. C. Campbell and J. L. Fowler, Bull. Am. Phys. Soc. 5, 410 (1961).

^{(1960).}

¹⁰ The Isotopes Division of Oak Ridge National Laboratory, which separated the lead isotopes, gave the following analysis for the sample: Pb^{208} (99.75%±0.05%), Pb^{207} (0.05%), Pb^{208} (0.2%); a spectroscopic analysis indicated 0.01% sodium and set the upper limits on other possible elements.

¹¹ D. W. Miller in Fast Neutron Physics edited by J. B. Marion

 ¹⁴ D. W. Miller in *Fast Neutron Physics* edited by J. B. Marion and J. L. Fowler (Interscience Publishers, Inc., New York, 1962), Part II, pp. 986–1032.
 ¹² J. H. Gibbons and H. W. Newson in *Fast Neutron Physics* edited by J. B. Marion and J. L. Fowler (Interscience Publishers, Inc., New York, 1960), Part I, pp. 133–174.
 ¹³ J. D. Kington, J. K. Bair, H. O. Cohn, and H. B. Willard, Phys. Rev. 99, 1393 (1955).
 ¹⁴ F. D. Brooks, Nuclear Inst. and Methods 4, 151 (1959).
 ¹⁵ M. Forte. Proceedings of the Second United Nations Internet.

¹⁵ M. Forte, Proceedings of the Second United Nations Interna-tional Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958 (United Nations, Geneva, 1958), Vol. 14, p. 300.



FIG. 1. Total neutron cross section of Pb²⁰⁸ as a function of energy. Dotted portion of curve below 720 keV due to Duke University measurements (see reference 22).

neutron-detecting crystal was located in line with the direction of the proton beam and 51 cm from the lithium target. During the course of the measurements, the $1\frac{1}{8}$ -in.-diam samples were reworked several times because this separated isotope was being used also at Duke University. For most of the measurements the Pb²⁰⁸

sample thickness was 17.86 g/cm^2 or 18.10 g/cm^2 . In the experiment it was positioned halfway between the source and the detector. A remotely controlled sample changer allowed the measurements to be made with a minimum delay between the sample-in and the sample-out runs. Neutrons were monitored either with a long

En (MeV)



FIG. 2. Differential cross sections of normal lead as a function of energy and angle in center-of-mass system. The data at 1.2, 2.2, and 3.2 MeV were obtained with an energy spread of 50 keV (see reference 7).

counter at a backward angle or with a $1\frac{1}{2}$ -in.-diam stilbene crystal located at about 80° from the forward direction. Gamma-ray pulses from this crystal were also suppressed by use of a separate Forte circuit. Counts due to scattered neutrons (less than 1.5% of the sampleout counts) were found by placing a 28-cm-long lucite cone between the source and the detector. The data in Fig. 1 have been corrected for this background.

In evaluating the in-scattering correction it is necessary to have an estimate of the 0° differential cross section of $Pb^{208,11} \sigma_n(0^\circ)$. For this purpose the measured differential cross section of normal lead was used (Fig. 2). Since at 1.2, 2.2, and 3.2 MeV the

measured differential cross sections extrapolated to 0° of normal lead and of a sample of effective composition 72% Pb²⁰⁸ agree within their estimated error,⁷ it is expected that the average value of $\sigma_n(0^\circ)$, as obtained from Fig. 2, will not differ greatly from such an average cross section of Pb²⁰⁸. Use of the curves for normal lead allows one to include many other measurements published in the literature.^{16–20} Of course, this procedure

¹⁶ M. Walt and H. H. Barschall, Phys. Rev. 93, 1062 (1954.)

¹⁵ M. Walt and H. H. Barschall, Phys. Rev. **93**, 1002 (1954.)
 ¹⁷ W. J. Rhein, Phys. Rev. **98**, 1300 (1955).
 ¹⁸ H. R. Brugger, H. J. Gerber, B. Lüthy, and A. E. Remund, Helv. Phys. Acta **28**, 331 (1955).
 ¹⁹ W. D. Whitehead and S. C. Snowdon, Phys. Rev. **92**, 114 (1977).

(1953)

²⁰ M. Walt and J. R. Beyster, Phys. Rev. 98, 677 (1955).

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TABLE 1. Properties of prominent resonances.					
IV Peak	v	VI	VII Reduced	VIII width in	IX
height		$2\pi\lambda^2(2J+1)$	units $3\hbar^2/2ma^2$		of
(barns)	J	(barns)	+parity	-parity	assignment
11.1 ± 0.7	5/2	10.90	0.02(2)	0.12(3)	A
5.7 ± 0.6	3/2	6.84	0.02(2)	0.01(1)	В
9.3 ± 0.4	5/2	9.60	0.04(2)	0.19(3)	A
5.6 ± 0.6	3/2	6.15	0.02(2)	0.01(1)	В
5.7 ± 0.6	3/2	5.52	0.03(2)	0.01(1)	A
3.7 ± 0.6	3/2	4.73	0.02(2)	0.01(1)	В
4.3 ± 0.6	3/2	4.56	0.02(2)	0.01(1)	В
$1.9{\pm}0.4$	1/2	2.18	0.01(0)	0.01(1)	В
4.3 ± 0.4	3/2	4.24	0.03(2)	0.02(1)	A

0.03(2)

0.01(0)

0.01(0)

0.02(2) 0.01(2)

0.01(2) 0.01(2)

0.02(2)

0.01(2)

0.09(0) 0.15(2)

0.14(4)

0.06(4)0.01(2)

0.01(0)

 $0.00\dot{4}(1)$

4.09

1.99

1.94

3.79

3.67

5.34

3.29

3.24

1.61

4.64

1.53

3.07

6.01

5.97

2.93

1.40

m **T D** . •

3/2 1/2 1/2 3/2 3/2 5/2

 $\frac{3}{2}$ $\frac{3}{2}$

 $\frac{1/2}{5/2}$

1/2 3/2 7/2 7/2 3/2

1/2

 3.2 ± 0.5

 1.5 ± 0.4

 1.8 ± 0.4

 3.7 ± 0.4

 3.6 ± 0.6

 5.2 ± 0.5

 3.5 ± 0.5

 3.2 ± 0.5

 2.0 ± 0.5

 4.7 ± 0.6

 2.2 ± 0.6

 6.1 ± 0.5

 5.9 ± 0.7

 3.1 ± 0.7

 1.2 ± 0.4

of making the in-scattering correction for high-resolution data by use of low-resolution differential measurements allows only average in-scattering corrections to be applied. Since $\sigma_n(0^\circ)$ changes considerably as one goes over a resonance, the actual correction will vary about the mean correction (0.04-0.07 b over the energy)range of Fig. 1) in the same ratio as the actual $\sigma_n(0^\circ)$ varies about the average $\sigma_n(0^\circ)$. As soon as the highenergy resolution differential cross sections become available, the data in Fig. 1 can be recorrected for inscattering from the information given in this paper by use of Eq. 10 of reference 11. Preliminary differential cross section measurements at 723- and 821-keV resonances show that the additional in-scattering correction due to this effect is about +0.1 b at these resonances.²¹

III

г (keV)

 $5.0\pm$ 1.0

 5.0 ± 1.0 5.5 ± 1.0 10.0 ± 1.0 5.0 ± 1.0 8.0 ± 1.0 6.5 ± 1.0 7.5 ± 1.0

 7.5 ± 1.0

 12.0 ± 1.5

 12.0 ± 2.0

 7.0 ± 1.0

 5.0 ± 1.0

 9.5 ± 1.5

 $3.0\pm$ 1.0

 6.5 ± 1.0

 6.5 ± 1.0

 $4.0\pm$ 1.0

 6.5 ± 1.0

 90 ± 10.0

 $10.5\pm~1.0$

 4.5 ± 1.0 3.0 ± 1.0

 13.0 ± 1.5

 $10.0\pm$ 1.0

II

 ΔE to $\pm 20\%$

(keV)

3.3

3.3 $3.2 \\ 3.2$

3.1 1.7 1.7

2.02.0

2.0

2.1

2.1

2.0

2.6

2.9

3.0

3.0

3.0

2.5

2.5

2.5 2.5 2.8

3.0

Ι

 E_0

(keV)

723

769

821 855 953

1112 1154

1204

1239

1285

1318

1354

1386

1431

1477

1600

1620

1632

1701

1715

1749

1761

1797

1872

The presence of a second lower energy group of neutrons from the $Li^7(p,n)Be^7$ source made it necessary to apply a further correction. This is proportional to the product of the following factors: the difference be tween the total neutron cross section at the energy of the primary group of neutrons and that at the lower energy group, the relative number of lower energy neutrons, and the relative efficiency of neutron detection for the two energies. Since the efficiency of the neutron detecting crystal as measured relative to that of a long counter was a rapidly increasing function of neutron energy, the effect of the second group of neutrons was considerably reduced. For example, during one typical run at 1500 keV, the stilbene crystal detected primary energy neutrons incident upon it with about 7%

efficiency; for the second group of neutrons at 1029 keV, this efficiency was only 0.9%. The data in Fig. 1 have been corrected for this second group of neutrons on the basis of the information compiled recently on the $Li^{7}(p,n)Be^{7}$ reaction.¹² The cross-section errors shown in Fig. 1 are calculated from the counting statistics. Points shown are, for the most part, for single runs; there were, however, many repeat runs which give one confidence in the data presented in Fig. 1.

0.02(1)

0.01(1)

0.01(1)

0.01(1)

0.03(3)

0.01(1)

0.01(1)

0.01(1)

0.02(3)

0.11(1)

0.04(3)

0.01(3)0.003(1)

0.01(1)

0.004(1)

DISCUSSION OF RESULTS

In Fig. 1 the total cross section of Pb²⁰⁸ is plotted as a function of neutron energy from 720 to 1890 keV. This corresponds to an excitation energy in Pb²⁰⁹ from 4.65 to 5.81 MeV. A number of lithium targets of different thicknesses were used in accumulating these data. The energy resolution for various portions of these curves can be inferred from Table I, which gives this information for prominent resonances. Recent measurements at Duke University of the total cross section of Pb²⁰⁸ (by use of the same enriched sample) have been made up to 732 keV.²² These measurements, which show the resonance at 723 keV, are in good agreement with the data reported here in the region of overlap.

From Fig. 1 one finds there are at least 85 neutron resonances in a 1170-keV energy range. These resonances have peak heights from which one deduces total angular momentum quantum number ranging from J=1/2 to J=7/2 (see Table I). This detailed structure,

 $_B^A$

В

A

A

В

A

A

A

В

A

В

A B

B

В

²¹ J. L. Fowler, Program of the Southeastern Section Meeting of the American Physical Society, Abstract 13, 1962.

²² G. C. Kyker, Jr., E. G. Bilpuch, J. A. Farrell, and H. W. Newson, Bull. Am. Phys. Soc. 7, 289 (1962); and E. G. Bilpuch (private communication).

also indicated by the 10 keV resolution data,³ was not expected for the closed shell plus one nucleus, Pb²⁰⁹.

In Table I is collected information on those prominent resonances having a natural width equal to or greater than the experimental resolution, which are sufficiently separated from neighboring levels so that reasonably definite conclusions can be drawn with regard to their resonance parameters. Column I gives the energy at the peak of the resonance. Column II gives the experimental energy resolution, estimated to be good to $\sim \pm 20\%$. The total width of the resonance at one-half maximum is given in column III together with an estimate of the errors. This total width, Γ , has been corrected for the effect of the experimental resolution. In column IV is recorded the value of the peak cross section minus the nonresonant contribution to the cross section. This column is corrected for energy resolution²³ under the assumption that the resolution function is Gaussian. The estimated uncertainty is based on statistics of the data indicated in Fig. 1, the uncertainty of the nonresonant cross section, and the uncertainty of the correction for the energy resolution arising from errors in the ratio $\Delta E/\Gamma$. Columns V and VI give the *J*-value assignment and the theoretical peak cross section, $2\pi\lambda^2(2J+1)$, expected for this assignment, where λ is the neutron wavelength divided by 2π . In most cases the estimated error in the peak cross section overlaps the theoretical cross section and is sufficiently small so as to distinguish the assigned J value from $J \pm 1$. From the quality of the fit as well as from a judgment as to how well the resonance was resolved, the reliability of the assignments is indicated by the notation in column IX. A signifies a resonably certain assignment; B a tentative assignment.

Columns VII and VIII of Table I give an estimate of the reduced width in units of $3\hbar^2/2ma^2$ under the two possible parity assignments for the resonance. Here \hbar is Planck's constant divided by 2π , m is the reduced mass of the neutron, and a the effective radius of interaction. This effective radius, a, was chosen in the following manner: For g waves (l=4), the highest possible orbital angular momentum allowed for a J = 7/2resonance, a phenomenological potential similar to that proposed by Blomqvist and Wahlborn²⁴ was used to calculate the width of a single-particle $g_{7/2}$ state at resonant energy by a method previously described in the case of O¹⁶ scattering.^{2,3} Blomqvist and Wahlborn choose a Woods-Saxon potential, $V = -V_0 \{1 + \exp i \}$ [(r-R)/A]⁻¹, with a diffuseness, A = 0.67 F. They take the $4s_{1/2}$ single-particle state in Pb²⁰⁹, before it is perturbed by a quadrupole oscillation, to be at 1.22 MeV below the energy of Pb²⁰⁸ plus a free neutron of zero energy. Combining this information with a knowledge of the $s_{1/2}$ scattering phase shifts as determined by Wilenzick et al.,⁸ one finds R=7.64 F, compared to 7.52 F chosen by Blomqvist and Wahlborn.

With these nuclear size parameters and a suitable choice of the potential depth and Thomas spin-orbit term, one calculates the width of a single-particle $g_{7/2}$ state at a laboratory energy of 1749 keV, the energy of one of the 7/2 resonances.^{3,25} This calculation gave the width at resonance energy to be ~ 80 keV. The radius of interaction a=9.3 F, used in the Wigner resonance formalism,²³ was chosen such that the penetration factor for l=4 waves²⁶ reduced the $3\hbar^2/2ma^2$ estimate of a single-particle width to approximately this value. Since the penetration factor for partial waves with l < 4 is less sensitive to the radius of interaction than for waves of l=4, this procedure permitted a reasonable first approximation to the estimate of the reduced width. Also given in parentheses in these columns is the assumed l value beside the reduced width to which it applies. Except in the case in which the J value is uncertain, this information is redundant in that the parity under which the reduced widths are listed together with J gives the orbital angular momentum directly.

For the 723-keV resonance, the energy, width, and cross section of which has been confirmed at Duke University,²² the observed peak height agrees well with that expected for a J=5/2 resonance. This result is in disagreement with the assignment^{6,8} J=3/2 made on the basis of information obtained with radiogenic and normal lead. The present 3-keV resolution data have shown that several peaks which at 10-keV resolution³ appeared to be due to single resonances, such as one at 830 keV, are, in fact, averages over several resonances so that spin assignments made on the single resonance hypothesis are without foundation. The J-value assignment of the broad resonance at 1.72 MeV, upon which a number of resonances are superimposed, is uncertain. It could be 3/2 as suggested earlier,⁷ but J=1/2 is also a possible assignment. It must be pointed out that even with 3-keV resolution, a number of resonances are not resolved, so that it is probable that some of the resonances listed in Table I may be in fact averages over several narrow resonances.

It is evident from an examination of the reduced widths in Table I that most of the resonances arose from compound nucleus effects. These estimates of the reduced widths allow one to draw conclusions with regard to the parity of the 723- and the 821-keV resonances. If these were $f_{5/2}$ resonances they would have a fair fraction of a single-particle width. Since no $f_{5/2}$ single-particle resonance is expected in this energy region,²⁴ one concludes that these are not f resonances. Thus, one makes a tentative assignment J=5/2+ for the spin and parity of these resonances. Differential

²³ H. B. Willard, L. C. Biedenharn, P. Huber, and E. Baum-gartner, in Fast Neutron Physics, edited by J. B. Marion and J. L. Fowler (Interscience Publishers, Inc., New York, 1962), Part II, pp. 1217–1316.
 ²⁴ J. Blomqvist and S. Wahlborn, Arkiv Fysik 16, 545 (1960).

²⁶ Mary Jo Mader and J. L. Fowler, Tenn. Acad. Sci. 37, 70 (Abstract) (1962).

J. E. Monahan, L. C. Biedenharn, and J. P. Schiffer, Argonne National Laboratory Report ANL-5846, 1958 (unpublished).

cross section measurements confirm this assignment.²¹ The asymmetry of these resonances about their resonant energy could arise from interference of resonant scattering with *d*-wave potential scattering.

This type of analysis does not fully apply for the J=7/2 resonances at 1749 and 1761 keV. These resonances could be associated with the $2g_{7/2}$ member of the 2g single-particle states, the lower member of which is $2g_{9/2}$ ground state of $\mathrm{Pb}^{209}\!.$ On the basis of recent measurements of (d,p) stripping on Pb²⁰⁸,²⁷ and reinterpretation of earlier data²⁸ by use of improved stripping theory calculations,²⁹ the $2g_{7/2}$ single-particle state has been identified as a bound-state level at an excitation energy of 2.47 MeV above the $2g_{9/2}$ ground state of Pb²⁰⁹. This assignment makes it improbable that there are $g_{7/2}$ levels at an excitation energy of 5.67 MeV (corresponding to resonances near 1750 keV) which have such a large fraction of a single-particle reduced width as indicated in Table I. Thus, one suspects that the 1749- and the 1761-keV resonances are $f_{7/2}$ resonances. This point can be settled more definitely by measurement of the neutron elastic differential cross sections since such measurements for a zero-spin target nucleus allow one to identify the parity of resonances.²³ Such differential cross sections, however, have to be measured with an energy spread not much larger than the width of the resonances. The high efficiency of

(1962). ²⁸ B. L. Cohen, R. E. Price, and S. Mayo, Nuclear Phys. 20, 370 (1960).

²⁹ R. H. Bassel, R. M. Drisko, and G. R. Satchler, Bull. Am. Phys. Soc. 6, 67 (1961).

crystal detectors used with Brooks-Owen techniques for gamma-ray suppression makes it possible to measure angular distributions with energy spread sufficiently small to resolve many of the Pb²⁰⁹ resonances.²¹

Although the recent identification of the singleparticle states of Pb²⁰⁹ seems satisfactory,^{27,30} this is such an important nucleus that there should be more confirming evidence for the level assignments. As pointed out by Satchler,²⁹ theoretical (d,p) stripping angular distributions even at 15 MeV are not particularly sensitive to *l*-values of the captured neutron for $l \ge 2$, a conclusion which is in agreement with experiment.^{27,28} At 20 MeV, however, theoretical (d, p)stripping angular distributions look considerably different for different l values of the captured neutron. This 20-MeV (d, p) stripping experiment would serve to confirm the information on the bound states necessary for the understanding of one of the simplest of the heavy nuclei. Such bound-state information is a prerequisite for a reasonably complete analysis of the virtual states of Pb²⁰⁹ which are observed in neutron total and differential cross section experiments.

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²⁷ Paresh Mukherjee and B. L. Cohen, Phys. Rev. 127, 1284

³⁰ J. R. Erskine and W. W. Buechner, Bull. Am. Phys. Soc. 4, 360 (1962).