central energy-production rate of a few thousand ger/g sec, a central temperature of about $T_c \approx 1.3$ is required (the effect of electron screening is negligible in this case).

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Neutron Total Cross Section of Np^{237} from 0.02 to 2.8 ev*†

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The total neutron cross section of Np^{237} has been measured in the energy region 0.02 to 2.8 ev with the Materials Testing Reactor crystal spectrometer. Measurements were made on a sample containing 152 mg of Np^{237} in oxide form dissolved in deuterated nitric acid. A value for the absorption cross section at 0.025 ev of 170±22 barns was obtained. Resonances were observed at energies 0.489±0.002, 1.337±0.015, and 1.488±0.018 ev, with the respective values of $\sigma_0\Gamma$ being 84.2 ev barns, approximately 29 ev barns, and approximately 140 ev barns. Values for σ_0 of 2600±100 barns, Γ of 0.032±0.003 ev, and $g\Gamma_n$ of 0.016 mv were determined for the 0.489-ev resonance.

I. INTRODUCTION

THE thermal-activation cross section of Np^{237} was determined by Jaffey and Magnusson[1] from radiometric measurements of the Np^{238} and Pu^{238} activities resulting from neutron capture in Np^{237}. A recent re-evaluation[2] of this experiment, in which a later value of the half-life of Pu^{238} was used, gave 170±20 barns for the activation cross section of Np^{237}. Since Np^{237} is used as a fast-neutron flux monitor in the presence of slow neutrons, a knowledge of the thermal neutron cross section is necessary to correct for the fission in the Pu^{238} formed. It is of interest to nuclear theory to know the level spacing and other parameters for this nucleus, which is one of the heaviest odd proton-even neutron nuclei available for such measurements.

The problems associated with transmission measurements on limited quantities of material have been treated by Shull and Wollan,[3] and by Bernstein *et al.*[4] An important consideration in such measurements is the arrangement of the sample in the thickest manner consistant with a beam sufficiently large to provide satisfactory counting statistics. The theoretical aspects of optimizing sample geometry have been considered by Rose and Shapiro.[5] With the high neutron flux provided by the Materials Testing Reactor the requirements for sample sizes are less critical, and transmission measurements may be extended to higher energies.

II. EXPERIMENTAL DETAILS

The spectrometer system consists of a collimator located in the reactor shielding, the spectrometer proper, a monochromating crystal, an automatic sample changer, slits to define the Bragg beam, a neutron detector, and the associated control and counting electronics. The neutron beam from the reactor was defined by a steel collimator of inside dimensions $\frac{1}{8}$ in.×$2\frac{3}{4}$ in.×96 in. This system, when the (240) planes of NaCl crystal were used, had an overall resolution of 0.9 μsec/meter. The (220) planes of NaCl were used to obtain some of the lower energy data.

The cross-section data reported here were obtained by transmission measurements on a sample of Np^{237} having a total sample cross section of 0.06 cm^2 at 0.025 ev. Since Np^{237} has a half-life of 2.20×10^6 years, the problems of radiolysis and alpha heating are not troublesome, and specialized handling equipment is not necessary. Since only 152 mg of Np were available for the measurements, a special sample changer was developed to allow the placement of the optimum amount of sample in the Bragg beam. For these measurements the Bragg beam was reduced to a width of 0.24 cm and a height of 1.5 cm by a boron carbide and paraffin collimator 12.7 cm long placed adjacent to the sample between the sample and the detector.

The detector was a group of three $B^{10}F_3$ proportional counters located with the cylindrical axes parallel to

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† A preliminary report of these measurements was made at the 1955 Washington, D. C. meeting of the American Physical Society [Phys. Rev. **99**, 611(A) (1955)].

[1] A. H. Jaffey and L. B. Magnusson, Atomic Energy Commission Report, ANL-4030, 1947 (unpublished).

[2] *Neutron Cross Sections*, compiled by D. J. Hughes and J. A. Harvey, Brookhaven National Laboratory Report BNL-325 (Superintendent of Documents, U. S. Government Printing Office, Washington, D. C., 1955).

[3] C. G. Shull and E. O. Wollan, Phys. Rev. **81**, 527 (1951).

[4] Bernstein, Borst, Stanford, Stephenson, and Dial, Phys. Rev. **87**, 487 (1952).

[5] M. E. Rose and M. M. Shapiro, Phys. Rev. **74**, 1853 (1948).

the Bragg beam. The active volumes of the counters were 18 in. long and 1 in. in diameter. The counters had 0.006-in. diameter center wires and were filled to a pressure of 65 cm of Hg. The counting rate at 0.025 ev for a Bragg beam area of 0.36 cm^2 was approximately 10^3 counts per second. The automatic-control and counting circuitry of the spectrometer has been described elsewhere.[6]

The fabrication of limited quantities of material into small uniformly thick samples is difficult, and it is usually expedient to dissolve the target material in a solution of deuterated mineral acid. The use of light hydrogen acids is undesirable because of their higher attenuation of the neutron beam. The neptunium used in these measurements was prepared, purified, and placed in DNO_3 solution by Dr. James Wallmann of the University of California. Final steps in the chemistry consisted of a conversion of the neptunium to Np_3O_8 and dissolution of the oxide in 2.91N DNO_3.

Two solutions of different concentration were prepared. One, containing 60.8 mg of Np per ml, was used for measurements in the region of major resonances; and the other, containing 304 mg of Np per ml, was used for measurements in the valleys between resonances and in the energy region below 0.4 ev.

The "thick sample," consisting of 500±1 microliters of the 304-mg/ml solution, was contained in a quartz cell having the inside dimensions 3.0×9.0×20 mm. The cell was closed with a fitted ground stopper and Glyptal was applied to the edges of the seal to prevent creeping of the material and possible changes in concentration through evaporation. A matched cell was filled with 2.91N DNO_3. The ratio of the counts obtained with the two cells placed alternately in the Bragg beam was approximately the transmission of the Np_3O_8. The cells were mounted in the vertical automatic sample changer. Horizontal and vertical alignment with respect to the Bragg beam collimator was accomplished by means of set-screw adjustments.

The "thin sample" was prepared by placing 400 microliters of the "thick-sample" solution in a quartz Beckman spectrophotometer cell and diluting it with D_2O to a volume of 2 ml. This sample was large enough to utilize most of the Bragg beam. A suitable blank was prepared by filling a matched cell with equivalent quantities of D_2O and DNO_3. These cells were mounted on a turntable designed to move the cells in and out of the beam in an alternate manner.

The neptunium concentration was determined by a radiometric assay. A 25-microliter aliquot of the thick-sample solution was diluted to a volume of 10 ml. Aliquots of 100 microliters of this solution were evaporated on platinum plates and were heated to red heat in an induction heater to remove the trace quantities of volatile matter. No sample localization effects were detected by a microscopic examination. Since the maximum amount of mass occurring as nonvolatile material on the plates was less than 0.01 microgram, the effects of self-absorption of alpha particles were regarded as negligible. The plates were counted with 2π geometry in a gas-flow proportional counter calibrated against samples of RaF standardized by the National Bureau of Standards.

Spectrographic analysis[7] revealed the presence of the following impurities: Al, 0.1%; Ca, 0.07%; Fe, 0.07%; Mg, 0.03%; and U, 3.0%, where the percentages are relative to the weight of Np_3O_8. An additional analysis[8] revealed the presence of 0.13% boron. With the exception of boron and uranium these impurities had no appreciable effect on the cross section measurements, and corrections for the effects of these two elements were applied to the data. A small contamination of Pu^{239} and Am^{241} detected through alpha pulse-height analysis[9] necessitated a 0.5% correction to the assay data to account for alpha events arising from these impurities.

III. RESULTS

The total cross section of Np^{237} as a function of energy is shown in Fig. 1. Major resonance peaks occur at 0.489±0.0017, 1.337±0.015, and 1.488±0.018 ev; and there is some evidence that small peaks exist at higher energies. Positive identification of the smaller peaks is made difficult by the closeness of the observed widths

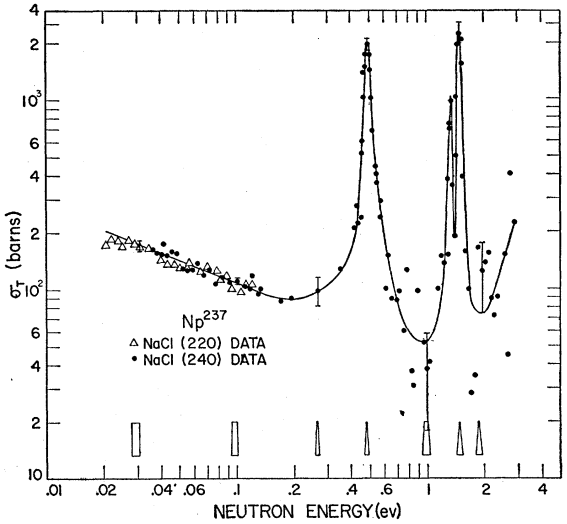

FIG. 1. Neptunium-237 total cross section. The indicated precision limits are those due to counting statistics only. The half-width of the triangles represents the full width at half maximum of the resolution function of the crystal spectrometer and the spacing between the high- and low-energy values of the individual data which have been averaged for convenience in plotting.

[6] G. L. Smith and L. G. Miller, Atomic Energy Commission Report, IDO-16168, 1955 (unpublished).

[7] James Wallmann (private communication).
[8] G. V. Wheeler (private communication).
[9] A. Ghiorso (private communication).

to the resolution width of the spectrometer and by the high transmission of the sample in these regions. Data were taken from 0.02 to 0.12 ev with the (220) planes of NaCl. Below 0.030 ev the data were corrected for the effects of second order neutrons in the Bragg beam. In the region from 0.035 to 3.0 ev data were taken with the (240) planes of NaCl, and in the region of overlap with (220) plane data good agreement was found.

The data from 0.020 to 0.17 ev vary approximately as $1/v$ and give a value for the total cross section at 0.025 ev of 180 ± 22 barns. The limits are standard deviations which take into consideration the statistical accuracy of the transmission measurements, the uncertainties in the neptunium concentration, as well as uncertainties in the corrections applied for impurities and for differences in the "sample" and "blank" solutions. The subtraction of 10 barns, obtained from the formula for the geometrical scattering cross section, $4\pi(1.45\times10^{-13}A^{\frac{1}{3}})^2$ (cm^2) from the total cross section at 0.025 ev results in a value 170 ± 22 barns for thermal neutron absorption cross section.

Breit-Wigner parameters, σ_0 and Γ, were determined for the 0.489-ev resonance by two different methods. The resonance-fitting method described by Sailor,[10] which includes corrections for the instrument resolution and Doppler broadening, gave values of $\sigma_0=2600\pm100$ b, $\Gamma=0.032\pm0.003$ ev, and $g\Gamma_n=0.016$ mv. Since the nuclear spin of Np^{237} is $\frac{5}{2}$, the statistical weight factor, g, can have values of 5/12 or 7/12, and Γ_n is either 0.0384 mv or 0.0274 mv. Since this method was derived with the assumption that $E_0>0.5$ ev, its applicability to this resonance was uncertain. A two-sample area analysis[11] was made, which resulted in a value of $\sigma_0\Gamma=83.5$ ev barns in agreement with $\sigma_0\Gamma=83.2$ ev barns obtained from the values of the parameters listed above. Values of $\sigma_0\Gamma$, obtained by area analysis, for the 1.34- and 1.49-ev resonances are approximately 29 and 140 ev barns, respectively. Since $\Gamma_\gamma\approx\Gamma$, it is possible to obtain a value for Γ_n from the product $\sigma_0\Gamma$. For the 1.34-ev resonance, Γ_n is approximately 0.036 or 0.025 mv for the respective values 5/12 and 7/12 of the statistical weight factor.

[10] V. L. Sailor, Phys. Rev. **91**, 53 (1953).

[11] Seidl, Hughes, Palevsky, Levin, Kato, and Sjöstrand, Phys. Rev. **95**, 476 (1954).

IV. CONCLUSIONS

The average level spacing $\bar{S}$ for Np^{237} is one of the lowest yet observed. For the three low-energy levels $\bar{S}=0.5$ ev (the average spacing per spin state $D=1.0$ ev). The observed $\bar{\Gamma}_n^0/D$ is the order of 0.7×10^{-4}. The measurements reported here agree, in general, with the resonance structure reported by Adamchuck *et al.*,[12] but show a much smaller cross section at 0.025 ev. The thermal neutron absorption cross section of 170 ± 22 barns obtained in this experiment agrees with recently reported thermal activation cross-section values of 172 ± 7 barns obtained by Brown and Hall[13] and 169 ± 8 barns by Smith *et al.*[14] Since the correction necessary for the presence of boron involves an uncertainty of ±20 barns at 0.025 ev, the good agreement of the thermal neutron absorption cross section with other results is fortuitous.

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[12] Adamchuk, Gerasimov, Yefimov, Zenkevich, Mostovoi, Pevzner, Chernyshov, and Tsitovich, *International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1955* (United Nations, New York, 1956), Vol. 4, p. 216, Paper No. A/CONF.8/P/645, USSR.

[13] F. Brown and G. R. Hall, J. Inorg. Nuclear Chem. **2**, 205 (1956).

[14] Smith, Passell, Alley, and Lewis, Atomic Energy Commission Report, IDO-16225 (unpublished).