

using different telescope arrangements and counters of different efficiencies.

At any rate, conclusion (i) is independent of detailed assumptions and may be regarded as the principal result of this preliminary work.

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² M. G. E. Cosyns, *Bull. Acad. Belg.* **5**, 498 (1937).

³ F. L. Hereford, *Phys. Rev.* **75**, 923 (1949).

** More precisely

$$\eta = \frac{(AB'C) \cdot (ABC) - (AB'C) \cdot (ABC) \cdot (ACG)}{[(AB'C) \cdot (ABC) - (AB'C) \cdot (ABC) \cdot (ACG)] + [(ABC) - (ABC) \cdot (ACG)]}$$

*** All errors quoted are standard errors.

⁴ Gangnes, Jenkins, and Van Allen, *Phys. Rev.* **75**, 57 (1949).

⁵ Fraser, Ostrander, Van Allen, *Echo Lake Conference* (June 1949).

⁶ F. L. Hereford, *Phys. Rev.* **74**, 579 (1948).

⁷ E. P. Ney, *Echo Lake Conference* (June 1949).

⁸ Freier, Lofgren, Ney, and Oppenheimer, *Phys. Rev.* **74**, 1818 (1948).

Spontaneous Neutron Emission from Uranium and Samarium

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EXPERIMENTS with 200-micron Ilford C2 plates show that about 15 mg of B^{10} and 2 mg of Li^6 can be incorporated per ml of emulsion after soaking for 2 hours in a solution containing 10 g of lithium borate and 3 ml of glycerine per 100 ml without causing appreciable loss in sensitivity for densely ionizing particles.^{1,2} The high concentration of B^{10} and Li^6 nuclei, coupled with the large cross section for slow neutron capture, provide an exceedingly sensitive recording medium for the measurement of slow neutron flux. Thus, a detector plate situated 1.3 cm from a ~ 4 -mc Po-Be disk enclosed in the paraffin cylinder described in Fig. 1 recorded a population of 8.38×10^3 tracks per ml of emulsion originating from the $B^{10}(n, \alpha)Li^7$ reaction after only a 15-min. exposure. Counts of proton recoil tracks show that the source emitted a total of 3.85×10^6 fast neutrons during the same interval, indicating the registration of 1 ($B^{10}+n$)-track per ml of emulsion per 460 ± 24 fast neutrons generated.

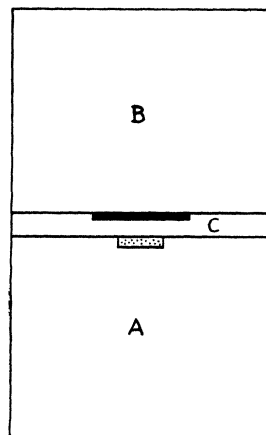


FIG. 1. Paraffin cylinder for slow neutron measurement. Two 1-lb. iron tobacco cans are each filled flush with 1450 g of molten paraffin. *A* serves as a sample holder and *B* as a neutron reflector. The plate holder *C* is a paraffin disk 16 mm thick carrying a 1×2-inch plate wrapped in paraffined-black paper. The cylindrical arrangement was chosen for convenience, and its efficiency has not been compared with a cubical or spherical disposition.

TABLE I. ($B^{10}+n$)-track counts from U and Sm.

Experiment	Exposure hours	Volume surveyed	Track count	Counts per ml per day
A—Blank	449	0.0155 ml	19	66
B—36.4 g U	449	0.0216	111	276
C—18.6 g Sm_2O_3 powder	449	0.0126	16	68
D—18.6 g Sm_2O_3 pellet	689	0.0080	21	92
E—20 g rare earth oxide pellet	689	0.0089	16	63
F—31.8 g U metal	689	0.0085	55	227

The calibration data suggest that by extending the exposure to 500 hours flux of the order of 2×10^{-6} slow neutron per sec. per cm^2 could be measured with an uncertainty less than 20 percent. As a test, three identical paraffin cylinders were assembled. *A*—containing paraffin only served as a control for cosmic-ray neutrons, *B*—contained a disk of uranium of approximately the same volume as the Po-Be source, and in *C*—a 25-ml cavity was filled with Sm_2O_3 powder. The units were stored in the sub-basement of a sea level building in which prior measurements showed an evaporation rate of 0.29 star per ml per day.³ The ($B^{10}+n$) tracks were counted with a $98\times$ fluorite objective and $7.5\times$ compensating oculars. High magnification is essential in order to discern the short tracks of 6- to 7-micron length at all angles of incidence and to differentiate them from occasional rod-shaped artifacts in the gelatin. The alpha-triton tracks from the $Li^6(n, \alpha)H^3$ reaction were noted, but were too few in number to constitute a quantitative check for the boron slow neutron reaction.

The differential counts between *B* and *A* (Table I) indicate the emission of 2660 fast neutrons per day per g of U. If this be attributed to spontaneous fission of U^{238} with one neutron released per fission, the count corresponds to a half-life of 1.8×10^{15} yr. This is in good agreement with the value of $(2.5 \pm 0.6) \times 10^{15}$ yr. observed by Maurer and Pose⁴ employing a cylinder of uranium weighing 8.82 kg.

As a check on the reproducibility of the method, the experiment was repeated with a smaller mass of uranium and an extension of exposure time. Also the samarium oxide was compressed into a pellet of 10-ml volume to provide better geometry, and a similar pellet of mixed rare earth oxides from Norwegian gadolinite was employed as a monitor. This measurement showed the emission of 2370 fast neutrons per day per g of U corresponding to a half-life of 2.0×10^{15} yr. The deviation from the mean value of $(1.9 \pm 0.1) \times 10^{15}$ yr. indicates a reproducibility of about 5 percent.

Compression of the samarium oxide powder increased the track count by 39 percent. If this be attributed to fast neutron emission from the samarium preparation, the half-life of the process is estimated at 10^{16} yr. In view of the small differential count and that neutrons may have originated by interaction of samarium alpha-particles with light nuclei impurities in our preparation, confirmation of the effect must be made using larger quantities of samarium specifically purified from beryllium, boron, and other elements with a large cross section for (α, n) interaction. The observed neutron production, if attributed to $Be^9(\alpha, n)C^{12}$, would necessitate the presence of about 1 percent BeO as impurity in the Sm_2O_3 . This degree of contamination is improbable in a rare earth preparation isolated from monazite sand.

Estimate of cosmic-ray neutron contribution to observed neutron emission from uranium.—An analysis of one of the $Li_2B_4O_7$ -loaded plates from the same batch employed in the measurements revealed a B^{10} concentration of 9.3×10^{20} atoms per ml. The plate exposed in the blank cylinder (*A*) exhibited 66 tracks per ml per day arising from the $B^{10}(n, \alpha)Li^7$ reaction. This affords a measure of the average cosmic-ray slow neutron flux in our laboratory during the period of the exposures:

$$\text{slow neutron flux} = \frac{66}{3525 \times 10^{-24} \times 9.3 \times 10^{20}} = 20.1 \text{ cm}^{-2}/\text{day}.$$

This value permits an estimate of the induced fission rate assuming 2×10^{-24} cm² for the fission cross section of normal isotopic uranium. In a mass of 36.4 g of the metal the number of induced fissions per day will be approximately:

$$20.1 \times 9.25 \times 10^{22} \times 2 \times 10^{-24} = 3.71.$$

If each fission releases between 1 and 3 fast neutrons, about 4 to 11 fast neutrons will be generated in the test mass of uranium per day of exposure. Since the plate records one track for each 460 fast neutrons, the correction seems too small for consideration.

Neutron yield from Sm-alpha-particles interaction with Be⁹.—The average cross section of the Be⁹(α ,n)C¹² reaction was approximated from the relationship $\sigma = \pi R^2 \bar{G}$. The average Gamow factor $\bar{G}$ for alpha-particle energies between 2.1 and 0 Mev is estimated at 0.067 using the Mattauch and Fluegge tables.

$$\sigma = \pi (2 \times 10^{-13} Z)^2 \times 0.067 = 0.021 \times 10^{-24} \text{ cm}^2.$$

The 18.6 g Sm₂O₃ emit:

$$18.6 \times \frac{300}{348} \times 89 \times 3600 \times 24 = 1.23 \times 10^8 \text{ alpha-particles per day.}$$

The differential count between Experiments D and E = 92 - 63 = 29 boron tracks per cc per day corresponds to the emission of $29 \times 460 = 13.3 \times 10^3$ fast neutrons. If these be attributed to (α ,n) on Be⁹, the number of the latter must be:

$$\frac{13.3 \times 10^3}{0.021 \times 10^{-24} \times 1.23 \times 10^8} = 5.16 \times 10^{21} \text{ Be}^9 \text{ atoms in 18.6 g of Sm}_2\text{O}_3$$

equivalent to a concentration of 0.37 percent Be or 1 percent BeO.

It is unlikely that our Sm₂O₃ pellet contains as much as 1 percent BeO. However, the additive effect of traces of Be, B, Mg, and Al may contribute in part to the observed neutron emission.

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Nuclear Spin and Quadrupole Coupling of S³⁵*

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THE rotational absorption transition $J=1 \rightarrow 2$ of carbonyl sulfide, OCS, containing S³⁵ has been observed at 23461 ± 5 mc/sec. This frequency agrees with the value calculated from bond distances and isotopic masses given in the literature, and directly from interpolation of the published frequencies of O¹⁶C¹²S³², O¹⁶C¹²S³³, O¹⁶C¹²S³⁴, and O¹⁶C¹²S³⁶.¹⁻³

Complete resolution of the spectrum was not attained. However, sufficient resolution of groups of hyperfine components was realized to permit a determination of the spin of S³⁵, the sign of the quadrupole coupling and an estimate of the magnitude of the coupling constant.

A typical record of the $J=1 \rightarrow 2$ transition with its associated hyperfine and Stark structure is shown in Fig. 1. Immediately below the experimental curve is the theoretical hyperfine structure pattern expected for a nuclear spin of $\frac{3}{2}$. All Stark components give negative deflections due to the properties of the phase-sensitive detector used. Comparison of our experimental results with theoretical patterns that would be expected for higher nuclear spins³ indicates that our results are not compatible with any spin value other than $\frac{3}{2}$. The frequency separation between the strong central components and the weaker components are 5.0 and 3.8 mc/sec., with the larger frequency separation on the high frequency side of the central components. The ratio of these



FIG. 1. Above: the experimental curve showing the partially resolved spectrum of COS³⁵. Below: the theoretical pattern calculated for a spin of $3/2$ for S³⁵.

separations is approximately 1.3, which agrees within our experimental error with the theoretical value of 1.43 for a spin of $\frac{3}{2}$. Comparison of our curves obtained from S³⁵ and S³³ which also has a spin of $\frac{3}{2}$ show that the patterns are mirror images, indicating that the quadrupole moment in S³⁵ is positive, or opposite in sign to that of S³³. We have estimated the electric quadrupole coupling constant $eqQ^{4,5}$ as $+20 \pm 4$ mc/sec. The corresponding value of S³³ in OCS is -28.5 ± 0.7 mc/sec.

The principal error in the quadrupole interaction was due to the absolute error in determining the dispersion of the recorded spectrum. This was measured with a wave meter at many points, and compared with the observed, known spectra of COS³³ and the Stark pattern of COS³⁴.

The detection system was a Stark modulation spectroscope operating at 100 kc/sec. and used a phase-sensitive or "lock-in" detector which drove an ink recorder. The 2K33 Klystron used was swept in frequency by a low speed mechanical drive operated by a synchronous motor. The absorption cell, cooled by dry ice, was 14 feet of K-band wave guide in which the Stark electrode was supported by quartz spacers.

The frequency of the central component was measured with a wave meter.

The OCS³⁵ was prepared from H₂SO₄ obtained from Oak Ridge. The ratio of S³⁵ to S³² at the time of use was about 0.004 and because of gaseous impurities the concentration of the OCS³⁵