Total Internal Conversion Coefficient of the 23.9-keV Ground-State Transition in ^{119m}Sn†

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The total internal conversion coefficient of the 23.874-keV transition in ^{119m}Sn is found to be $\alpha = 5.13 \pm 0.15$, on the basis of γ- and x-ray intensity measurements with a high-resolution solid-state x-ray spectrometer. Consequently, the Mössbauer cross section for resonant absorption by ^{119}Sn is $(1.40 \pm 0.05) \times 10^{-18}$ cm^2.

I. INTRODUCTION

THE energy[1] of the ground-state transition in ^{119m}Sn is 23.874 ± 0.010 keV and lies very close to the energy of Sn K x rays[2] [$E(K\alpha_2) = 25.044$ keV] that are emitted following K-shell internal conversion of the 65.66-keV isomeric transition[3] (Fig. 1). The difficulty of resolving 23.9-keV γ rays from Sn K x rays has, until recently, stood in the way of a direct determination of the total internal conversion coefficient α of the transition,[3] and coincidence measurements had to be employed that necessarily entail considerable uncertainties.[4]

Tin-119m is widely used in Mössbauer-effect studies. The total internal conversion coefficient of the ground-state transition employed in recoilless-absorption experiments is of importance in interpreting the results of such measurements. In the presence of internal conversion, the cross section for resonant absorption to the first excited state, with the on-resonance value of the γ-ray energy, is

$$\sigma_0 = 2\pi \lambda\!\!\!^{-2} \frac{2I_u + 1}{2I_g + 1} \frac{1}{1+\alpha}, \qquad (1)$$

where $\lambda\!\!\!^{-}$ is the (reduced) wavelength of the γ ray, while I_u and I_g are the upper- and ground-state spins, respectively.[5–7] From observed transmission curves in Mössbauer experiments, the quantity $\sigma_0 f_a$ is usually obtained, where f_a is the fraction of recoilless γ rays absorbed in a crystalline absorber. If σ_0 can be calculated, the Debye-Waller factor $2w_a$ can be found from $f_a = e^{-2w_a}$, and hence, the Debye temperature Θ can be determined.

The recent development of high-resolution solid-state x-ray detectors has made it possible to resolve the 23.9-keV γ ray from Sn K x rays, and hence, to carry out a more direct determination of α. By definition, the total conversion coefficient is $\alpha = (N_0/N_\gamma) - 1$, where N_γ is the number of 23.9-keV photons and N_0, the number of ^{119m}Sn decays, in a given time. Now the 65.7-keV $M4$ transition is essentially wholly converted[8] [theoretical $\alpha_K(65.7) = 1667$], whence $N_0 = N_K[1 + (L+M+\cdots)/K]_{M4}$. It follows that

$$\alpha = \frac{N_x}{\omega_K N_\gamma}\left[1 + \frac{L+M+\cdots}{K}\right]_{M4} - 1. \qquad (2)$$

Here, ω_K is the K-shell fluorescence yield of Sn, and N_x/N_γ is the intensity ratio of Sn K x rays and 23.9-keV γ rays. The determination of α described in the present paper is based on relation (2), with the intensity ratio N_x/N_γ being measured in a high-resolution Si(Li) x-ray spectrometer.

II. EXPERIMENT

A. Spectrometer

Photon spectra were measured with a solid-state x-ray spectrometer constructed in this laboratory after a Lawrence Radiation Laboratory design.[9] The spectrometer consists of a cooled 3-mm-thick lithium-drifted silicon detector, a field-effect transistor preampli-

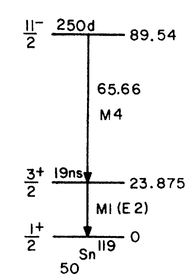

FIG. 1. Decay scheme of ^{119m}Sn, adapted from Ref. 3. Energies in keV.

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[1] J.-P. Bocquet, Y. Y. Chu. O. C. Kistner, M. L. Perlman, and G. T. Emery, Phys. Rev. Letters **17**, 809 (1966).

[2] J. A. Bearden, Rev. Mod. Phys. **39**, 78 (1967).

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[5] S. Margulies and J. R. Ehrman, Nucl. Instr. Methods **12**, 131 (1961).

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[7] G. A. Bykov and Pham Zuy Hien, Zh. Eksperim. i Teor. Fiz. **43**, 909 (1962) [English transl.: Soviet Phys.—JETP **16**, 646 (1963)].

[8] R. S. Hager and E. C. Seltzer, California Institute of Technology Report No. CALT-63-60, 1967 (unpublished).

[9] H. R. Bowman (private communication); E. Elad and M. Nakamura, Lawrence Radiation Laboratory Report No. UCRL-16515, 1965 (unpublished).

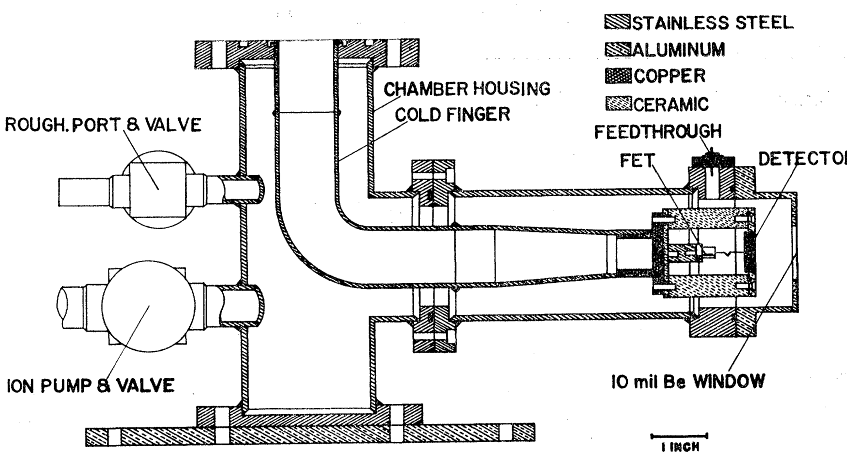

FIG. 2. Cross section of solid-state x-ray spectrometer.

fier operated at liquid-nitrogen temperature,[10] and a Tennelec TC-200 main amplifier. Detector and preamplifier are mounted in a stainless-steel vacuum chamber kept at 3×10^{-7} Torr by a 1-liter/sec Vacion pump and are cooled by liquid nitrogen from a Linde CR-10 cold-trap reservoir. A cross section of the device is shown in Fig. 2. Two ceramic rods provide a heat leak that keeps the detector near $-140°$C. The selected Texas Instruments 2N3823 preamplifier FET is mounted directly on the cold finger; a stainless-steel wire clip soldered to the gate lead presses the detector against its ring mount and provides contact. The spectrometer window is of 0.010-in. beryllium. Vacuum-tight ceramic feed-through insulators are used for detector bias and preamplifier output. Resolution slightly better than 1 keV full width at half-maximum (FWHM) is attained, as illustrated in Fig. 3.

The entire experiment was repeated with a second, similar spectrometer employing a 2-mm-thick detector, in order to guard against calibration and systematic errors.

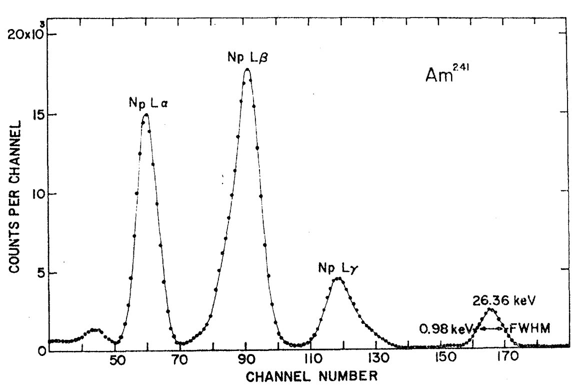

FIG. 3. Photon spectrum from ^{241}Am, illustrating performance of of Si(Li) x-ray spectrometer with cooled FET preamplifier.

[10] E. Elad, Nucl. Instr. Methods **37**, 327 (1965).

B. Calibration

Accurate determination of the efficiency of the Si(Li) detectors, as a function of x-ray energy, constitutes a crucial part of the experiment. The response curves were established with K x rays from a series of elements, excited by fluorescence. The relative intensities of these x rays, measured with the solid-state detector to be calibrated, were compared with relative intensities measured with a low-noise scintillation counter, using a highly reproducible geometry (Fig. 4). By choosing only x rays below the iodine K binding energy, escape-peak corrections for the scintillation counter were avoided. The measured relative efficiencies of each Si(Li) detector were least-squares fitted by computer to the functional form

$$\epsilon = ae^{-\mu b}(1-e^{-\mu c})\,, \qquad (3)$$

where a is the ratio of solid angles subtended by Si(Li) and scintillation detectors, $b=\rho x_D$ is the product of the mass density of Si and the thickness of the dead layer on the face of the diode, and $c=\rho x_S$ is the product of Si density and the depletion-layer thickness, while μ is the mass absorption coefficient of Si, at the weighted-average K x-ray energy. Figure 5 shows the efficiency curve for a "3-mm" detector, obtained in this manner.

C. Experimental Procedure

1. Analysis of Spectra

Tin-119 photon spectra were analyzed with the aid of a computer program that fits Gaussians of equal width above a straight-line background. In principle, the $K\alpha_1+K\alpha_2$ (25.271- and 25.044-keV) x-ray peak is slightly wider than the monoenergetic 23.874-keV γ-ray peak. However, this difference is negligible in the evaluation of intensity ratios in the actual spectrum, at a detector resolution of $\sim$1 keV FWHM. That the peaks can be fitted adequately with Gaussians of equal width was verified by constructing a mocked-up spectrum based on known x-ray energies and relative intensities; com-

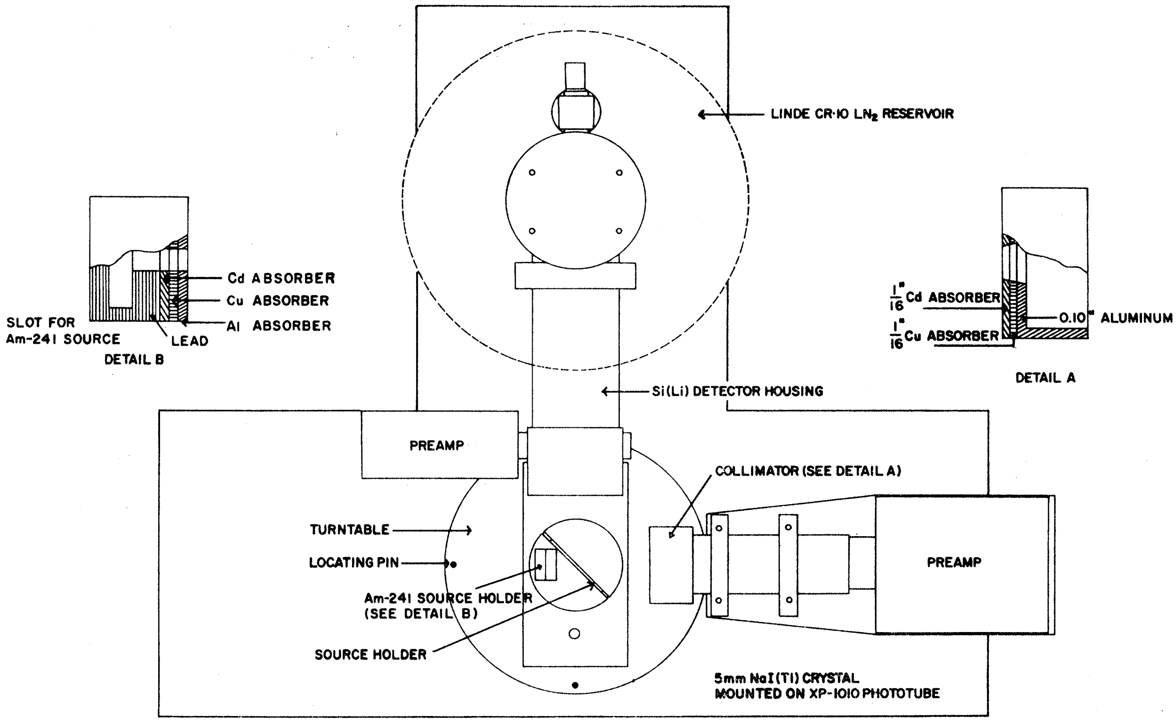

FIG. 4. Apparatus employed in the efficiency calibration of Si(Li) x-ray spectrometer.

puter analysis of the mocked-up spectrum yielded the input intensity ratios to better than 0.3%.

An alternative method of analysis was attempted. From ^{119m}Sn spectrum, a fluorescence-excited Sn x-ray spectrum was subtracted, normalized at the $K\beta$ peak. The difference between the two spectra then corresponds to the 23.9-keV γ ray. Less accuracy was attained with this subtraction method than with the computer analysis because of the need to employ exceedingly thin fluorescence sources, which produce low intensity and poor statistics. Thicker fluorescence sources radiate more strongly, but source thickness distorts the relative intensities of x-ray lines in the spectrum. Because of the strong dependence of the mass-absorption coefficient on energy, the Sn $K\beta/K\alpha$ intensity ratio, for example, is increased by 5% if the x rays traverse only 15 mg/cm^2 of Sn, and 66 mg/cm^2 of Sn produce a 25% enhancement of this ratio.

2. *Subtraction of In X Rays from ^{113}Sn Decay*

The ^{119m}Sn source material[11] contained a slight amount of ^{113}Sn impurity. It was necessary to correct the photon spectra for In K x rays emitted in ^{113}Sn decay. The amount of ^{113}Sn impurity was ascertained by measuring the intensity of the 393-keV γ ray (see Fig. 6) with a $1\frac{1}{2}\times 1$-in. NaI(Tl) scintillation detector of known efficiency.[12] The 393-keV γ-ray intensity was (2.05 ± 0.25)% of the x-ray and 23.9-keV γ-ray intensity at the time of assay; decay corrections were made for spectra obtained at other times. K-electron capture[13] of ^{113}Sn to the 393-keV level of ^{113}In results in 1.11 ± 0.03 In K x rays per 393-keV photon, and K-shell internal conversion of the 393-keV transition[14] pro-

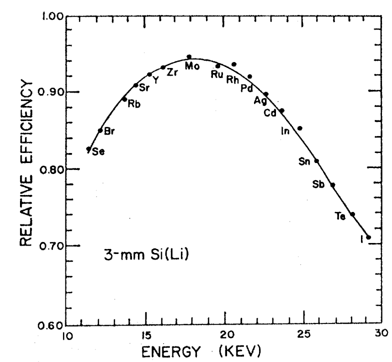

FIG. 5. Efficiency curve for Si(Li) x-ray detector of nominally 3-mm depletion depth. Dots represent experimental points, based on K x rays of the indicated elements; the curve is a least-squares fit of Eq. (3) of the text, with dead-layer thickness $x_D = 3.3\times 10^{-3}$ cm and effective depletion thickness $x_S = 0.34$ cm.

[11] Supplied by Nuclear Science and Engineering Co., Pittsburgh, Pa. 15236.

[12] Calculated scintillation detector efficiencies from M. I. Kalkstein and J. M. Hollander [University of California Radiation Laboratory Report No. UCRL-2764, 1954 (unpublished)], verified through measurements of ^{75}Se γ-ray spectrum.

[13] I. O. Durosinmi-Etti, D. R. Brundrit, and S. K. Sen, in *Internal Conversion Processes*, edited by J. H. Hamilton (Academic Press Inc., New York, 1966), p. 201.

[14] J. S. Geiger, in *Internal Conversion Processes*, edited by J. H. Hamilton (Academic Press Inc., New York, 1966), p. 379.

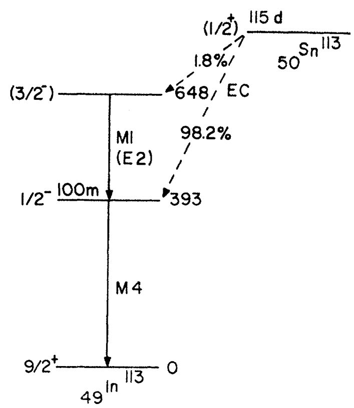

FIG. 6. Decay scheme of ^{113}Sn, adapted from Ref. 3. Energies in keV.

duces additional 0.38±0.01 In K x rays per 393-keV photon. Here, the K-shell fluorescence yield of In has

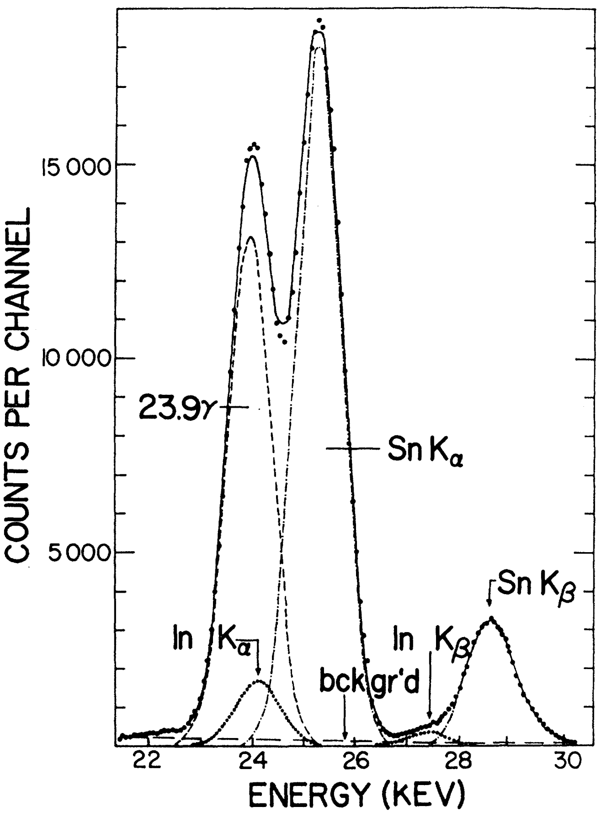

FIG. 7. Typical ^{119m}Sn photon spectrum, obtained with a Si(Li) x-ray spectrometer. Heavy dots represent experimental data. Two Gaussians of equal width plus a straight-line background have been computer-fitted to the data below 27 keV (solid line). The double peak is resolved into Gaussians due to the 23.9-keV γ ray, Sn $K\alpha$ x rays, and In $K\alpha$ x rays originating from a ^{113}Sn impurity in the source; the intensity of the In K x rays has been calculated from that of the 393-keV ^{113}Sn γ ray (see text). Above 27 keV, the solid curve simply links the data points. On the low-energy side of the Sn $K\beta$ x-ray peak, the contribution due to In $K\beta$ x rays is indicated. Asymmetry of the Sn $K\beta$ peak on the high-energy side is due to $K\beta_2$ x rays, which account for approximately 10% of the total $K\beta$ intensity and exceed the $K\beta_1$ x rays by approximately 600 eV in energy.

been taken as 0.83±0.01 after Fink *et al.*[15] and electron capture to the 648-keV level of ^{113}In has been neglected.

On the basis of these estimates, In $K\alpha$ and $K\beta$ x-ray peaks of appropriate intensities were subtracted in the analysis of the Sn photon spectra. The $K\alpha/K\beta$ intensity ratio was taken to be 4.80±0.02, after the tables of Storm and Israel[16] and of Wapstra *et al.*[17] A typical spectrum, resolved into its components, is shown in Fig. 7.

III. RESULTS AND DISCUSSION

From the analysis of nine runs taken over a period of four months with the 3-mm-thick Si(Li) detector, the intensity ratio of ^{119m}Sn $K\alpha+K\beta$ x rays to the 23.9-keV photons was found to be $N_x/N_\gamma=1.66\pm0.03$.

By interpolation from the tables of Hager and Seltzer,[8] the K conversion coefficient for the 65.7-keV $M4$ transition (taken to be of pure multipole order) is $\alpha_K(65.7)=1667\pm10$. With experimental relative L, M, and N conversion-electron intensities, recently measured by Bocquet *et al.*,[18] normalized to Hager and Seltzer's[8] α_{L_3}, one finds $\alpha_{L+M+N}(65.7)=3505\pm22$, and hence, $[(L+M+\cdots)/K]_{M4}=2.10\pm0.02$. With $\omega_K=0.840\pm0.010$ for tin,[15] Eq. (2) then yields

$$\alpha=5.13\pm0.15$$

for the total internal conversion coefficient of the 23.9-keV ground-state transition in ^{119m}Sn.

A separate series of measurements with a 2-mm-thick detector of poorer efficiency and resolution give the result $\alpha=5.13\pm0.46$, consistent with the above.

While the present work was in progress, an independent estimate of α from internal conversion studies was completed by Bocquet *et al.*[18] Utilizing the sum of the 23.9-keV conversion-electron intensities, observed in a high-resolution magnetic spectrometer, and assuming that the L_1 internal conversion coefficient has the theoretical value of 3.75±0.07 after Hager and Seltzer,[8] these authors derive $\alpha=5.12\pm0.14$. The excellent agreement of the result of Bocquet *et al.*[18] with that obtained in the present work by a totally different method can be construed to confirm the accuracy of

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[16] E. Storm and H. I. Israel, Los Alamos Scientific Laboratory Report No. LA-3753, 1967 (unpublished).
[17] A. H. Wapstra, G. J. Nijgh, and R. Van Lieshout, *Nuclear Spectroscopy Tables* (North-Holland Publishing Co., Amsterdam, 1959), p. 81.
[18] J.-P. Bocquet, Y. Y. Chu, G. T. Emery, and M. L. Perlman, Phys. Rev. **167**, 1117 (1968). We are much indebted to these authors for communicating their results in advance of publication.

the pertinent theoretical conversion coefficients computed by Hager and Seltzer.[8]

With the new value for α, the Mössbauer cross section for resonant absorption [Eq. (1)] is $\sigma_0=(1.40\pm0.05)\times10^{-18}$ cm^2, 6% higher than listed in the standard reference tables of Muir *et al.*[19] on the basis of earlier data.

[19] A. H. Muir, Jr., K. J. Ando, and H. M. Coogan, *Mössbauer Effect Data Index 1958–1965* (Interscience Publishers, Inc., New York, 1966), p. 126.

ACKNOWLEDGMENTS

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Nuclear Pairing Energy of Transition Nucleus Pu^{240}†

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Fission-fragment angular distributions are measured for the fission of Pu^{239} induced by monoenergetic neutrons of energies 150, 400, 475, 550, 600, 700, 800, 900, 1000, 1100, 1200, 1350, and 1500 keV (energy resolution, ±25 keV). Theoretical calculations are utilized to determine the parameter K_0^2 at each excitation energy from the observed angular distributions. The values of K_0^2 rise sharply from a value of 5–6, for neutron energies less than or equal to 600 keV, to an approximately constant value of 13, for neutron energies in the range 1.3–2.0 MeV. The break in the K_0^2 curve occurs at about 2.2 MeV above the fission threshold, and is interpreted as the beginning of two-quasiparticle excitations of the highly deformed transition nucleus Pu^{240}. This result suggests that the pairing energy gap is increased for large nuclear deformations.

I. INTRODUCTION

The capture of a neutron in Pu^{239} produces an even-even fissioning nucleus. It is well known that Pu^{239} has a large thermal-neutron fission cross section. Studies with resonance neutrons have revealed two groups of fission resonances.[1] The fission widths of each group of resonances is reasonably well fitted with a Porter-Thomas distribution, indicating a single fission channel for each spin state.[1] Furthermore, from the measured average fission width, one deduces that the 0+ channel is completely open, whereas the 1+ channel is only partially open. The 0+ level is thought to define the fission threshold for an even-even transition nucleus. The above information suggests that thermal and resonance neutrons have insufficient energy to excite quasiparticle excitations in the transition nucleus Pu^{240}.

† Work supported by the U. S. Atomic Energy Commission.

[1] J. Blons, H. Derrien, A. Michaudon, P. Ribon, and G. de Saussure, Compt. Rend. **262**, 79 (1966); A. Michaudon, Second Conference on Neutron Cross Sections and Technology, 1968, Washington, D. C., Paper D1 (unpublished).

The present experiments were initiated to look for the two-quasiparticle threshold in the intermediate transition nucleus. Measurements of fission-fragment angular distributions as a function of energy reveal information on the transition states of a nucleus as it passes over the fission barrier.[2–6] These transition states

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[3] I. Halpern and V. M. Strutinsky, in *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958* (United Nations, Geneva, 1958), Vol. 15, p. 408.

[4] J. J. Griffin, in *Proceedings of the International Conference on Nuclear Structure, Kingston, Canada, 1960*, edited by D. A. Bromley and E. W. Vogt (The University of Toronto Press, Toronto, 1960), p. 843.

[5] J. A. Wheeler, in *Fast Neutron Physics, Part II*, edited by J. B. Marion and J. L. Fowler (Interscience Publishers, Inc., New York, 1963), p. 2051.

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