CONF-760715--1

LA-UR-76-1418

TITLE: SPIN DETERMINATION OF FISSION RESONANCES

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SUBMITTED TO: 1976 International Conference on the Interactions of Neutrons with Nuclei, Lowell, MA 01854, July 6-9, 1976

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ENERGY RESEARCH AND
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CONTRACT W-7405-ENG. 36

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SPIN DETERMINATION OF FISSION RESONANCES

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INTRODUCTION

Ten years ago, Eric Lynn noted[1] that understanding of the neutron cross sections of the non-fissile nuclei seemed fairly complete through measurements and complex potential models. He then explored the question of how far a similar program could be carried out for fissionable nuclei. Since the theory most fundamental to the understanding of fission cross sections at low neutron energies is the channel theory of A. Bohr,[2] Lynn examined the energies and nature of those internal nuclear states associated with the transition of the nucleus through the fission barrier, the fission channels. He concluded at that time that understanding of this basic theory was far from complete, if not even somewhat superficial, due largely to a lack of pertinent measurements. Unfortunately, although considerable effort has been expended to make relevant measurements and to pursue complex models, our understanding of the properties of the fission channels has been only little improved in the last ten years.

In the Bohr theory, the transition states or fission channels are collective in nature and characterized by the total angular momentum J, the parity π, and the projection K of J on the nuclear symmetry axis, assuming that the nuclear shape during the passing of the saddle point remains axially symmetric. These channels are assumed to occur in bands, corresponding to particular modes of collective motion, and each band is characterized by the same K value and parity. Within each band, there are a number of different J values. An open fission channel is one which is both energetically available and has the same J^{π} as the compound nucleus.

Probably the most straightforward approach to understanding the nature and the role of these fission channels is in the direct observation of slow neutron fission resonances. Ideally, one needs to determine the channel quantum numbers, J^{π} and K, the resonance parameters, E_0, Γ_n, Γ_f, and Γ_{γ}, and the detailed behavior of the fission products. In particular, one should study the prompt neutrons, the distribution of fragment masses and energies, and even the total neutron and γ-ray energies, for each fission resonance. Although broad in scope, these measurements are presently feasible for a variety of fissionable nuclei. Availability of intense pulsed neutron sources and, as we will show later, advances in cryogenic technology presently permits observation of the most elusive of these quantities, the channel quantum numbers. In this paper, we will examine both the present state of available information on the channel quantum numbers for resonance fission and the most urgently needed additional experiments. Although a wealth of information pertaining to resonance parameters in fissionable nuclei exists, very few measurements pertaining to channel quantum numbers have been made. The discussion in this paper will necessarily rely heavily upon the alignment measurements of Pattenden and Postma and upon the polarization results from an experimental program conducted jointly by Los Alamos and Oak Ridge scientists.

EXPERIMENTAL TECHNIQUES

A rather vast amount of effort has been expended in the past to determine the spins of fission resonances. Any detailed and complete discussion of the techniques employed would necessarily be lengthy. Briefly, the various techniques used may be divided into two basic classifications. The first includes the direct methods, which encompass both the polarization techniques where a polarized target and a polarized beam are used and the method of using the total and scattering cross sections to determine the statistical weight factor, g_J. The class of indirect methods includes all other techniques used to infer the spin of the fission resonances. Among those techniques are: 1) level interference effects in elastic scattering and radiative capture, 2) γ-ray transitions and multiplicities, 3) fission width distributions, 4) prompt neutron and γ-ray emission, 5) fission fragment mass asymmetry and kinetic energy distributions, and 6) the ratio of ternary to binary fission

Of the two direct methods, each has a disadvantage. The main disadvantage of the polarized target and polarized beam technique is its extreme complexity. Although the results are simple to interpret, i.e. resonances of one J value are diminished while the resonances of the other are enhanced, the experimental techniques and apparatus are formidable. Although this method is a virtually infallible method for distinguishing between s-wave resonances of different spin, care must be taken to determine the correct absolute spin values. The single method which has been successfully employed to produce significant polarization in fissionable targets uses the hyperfine splitting in ferromagnetic systems. This hyperfine field, which may be several MOe, interacts with the nuclear magnetic moment, μ, to produce the nuclear polarization. However, the sign of the magnetic moment is frequently unknown and the direction of the hyperfine field may be either parallel or antiparallel to the applied field. Usually, sufficient information exists, either from Mössbauer measurements or from model calculations, to determine the signs of μ and the hyperfine splitting. In addition, the behavior of the observed resonances, such as the spacing or width distribution, may permit absolute determination of spin values. A further indication of the absolute spin is found in the approximate expression relating the polarized cross section, σ_p, to the unpolarized cross section, σ_o:

$$\sigma_p = \sigma_o(1 + f_I f_N f_n) \qquad (1)$$

Here f_n is the polarization of the incident neutron beam, f_N is the polarization of the target, and f_I is a spin-dependent factor given by:

$$f_I = \frac{I}{I+1} \text{ for } J = I + 1/2$$

$$f_I = -1 \text{ for } J = I - 1/2.$$

Clearly, the enhancement or diminution of a resonance is greater for the $J = I - 1/2$ case. With sufficient nuclear polarization and with a reasonably low value of the target spin I, this distinction permits determination of the absolute value of J.

The method of using the total and scattering cross sections is difficult if $\Gamma n/\Gamma \ll 1$, a common occurrence for fissionable nuclei. Comparisons between spin assignments in ^{235}U[3] and in ^{237}Np[4] by the two direct methods show little better than random agreement, due presumably to the low ratio of Γ_n/Γ.

A general lack of consistent spin assignments for resonances in fissionable nuclei by the various indirect methods would by itself lead one to doubt these techniques. However, an excellent example for a detailed comparison between such assignments and those from a polarized beam and target experiment exists in the heavily studied system $^{235}U + n$. This comparison is detailed in Ref. 3 but we will review the basic results. Generally, agreement between the spin assignments from the indirect techniques and from the polarization experiments are nearly random with a single interesting exception. Four groups[5-8] of experimenters attempted to assign spins to low energy radiative capture resonances in ^{235}U by examining the de-excitation γ-rays. Three of these four measurements are in poor agreement with the polarization experiment while the work of Corvi et al[5] is in perfect agreement, for those limited cases which they studied. The remaining indirect techniques appear to be less fruitful, except in special cases such as ^{239}Pu where the ground state spin is 1/2. Only two K-bands exist, 0^+ and 1^+, with the 0^+ channel being fully open and the 1^+ only partially open.

The K-value of a fission resonance of known J may be directly determined by measuring the angular distribution of fission fragments from an aligned target. Such a measurement was attempted originally by Dabbs et al[9] and later by Pattenden and Postma[10] on ^{235}U in crystals of $UO_2\ Rb(NO_3)_2$, and by Kuiken et al[11,12] on ^{233}U and ^{237}Np in the same crystal. All these experiments were handicapped by the low thermal conductivity of the host crystals with a resultant low degree of alignment. This problem may be surmounted by using an intermetallic compound which exhibits antiferromagnetism. In such a compound, the relatively high thermal conductivity will permit one to reduce the temperature low enough to achieve a sufficient degree of alignment to unambiguously assign K-values to fission resonances. In principle, this measurement should be considerably simpler than the spin determination experiment which requires both a polarized beam and a polarized target.

EXPERIMENTAL RESULTS

Presently, unambiguous spin assignments for resonances in slow neutron-induced fission exist only for ^{235}U, ^{237}Np, and although somewhat more ambiguous, for ^{239}Pu.

Because of the scope and number of measurements on ^{235}U, the remainder of this paper will primarily address this single nucleus. We will examine the information available in the resolved region, say below 60 eV, and describe briefly the status of the unresolved region. In both regions the role of spin will be discussed, with help from some new results from an experiment using a polarized neutron beam and polarized ^{235}U target.

The results from a preliminary measurement made at the Oak Ridge Electron Linear Accelerator (ORELA) reported by Keyworth et al[3] in 1973 assigned spins to 65 resonances below 60 eV. The more recent measurement, with increased polarization and statistical accuracy, permit assignments to all known resonances in this energy region. In these measurements, the neutron beam was polarized by transmission through single crystals of $La_2Mg_3(NO_3)_{12} \cdot 24H_2O$ (LMN) in which the protons in the waters of hydration were dynamically polarized. The target was the intermetallic ferromagnetic compound US, which was polarized in a ^3He-^4He dilution refrigerator operated at $\sim 0.02°K$ and in a magnetic field ~ 5 kOe. The details of the methods used are described in Refs. 3 and 4.

The data consist of time-of-flight spectra of fission events occurring in the target with the neutron beam polarized parallel and antiparallel to the target polarization, and of the transmission under the same conditions. The degree of polarization of the neutron beam and of the target was approximately 50% and 15%, respectively. For the analysis of the more recent data, M. S. Moore has devised a new technique of separating the components of the cross section due to J = 3 and J = 4. If N_3 and N_4 are the J = 3 and J = 4 enhanced count rates, i.e. the spin antiparallel and parallel data, then we can write

$$N_3 = A_3\sigma_3\phi + A_4\sigma_4\phi \tag{2a}$$

and

$$N_4 = B_3\sigma_3\phi + B_4\sigma_4\phi. \tag{2b}$$

where σ_3 and σ_4 are the cross sections for J = 3 and J = 4, ϕ is the neutron flux, and the constants A_3, A_4, B_3, and B_4 are determined from f_H, f_N, and I as defined in Eq. (1). Solving for the appropriate spin-dependent components, we get

$$\sigma_3\phi = (B_4N_3 - A_4N_4)/(A_3B_4 - B_3A_4) \tag{3a}$$

and

$$\sigma_4\phi = (A_3N_4 - B_3N_3)/(A_3B_4 - B_3A_4) \tag{3b}$$

In Figs. 1-4 these quantities are plotted for the energy ranges 8-44 eV, where the resonances are resolved, and 200-260 eV, where the resonances are unresolved. This analytical technique has greatly facilitated the analysis in both regions. One can simply assign spins from examination of the plots. Using this technique, these recent data show clearly the existence of previously unresolved overlapping levels of different spin, as exemplified by the structure near 35 eV.

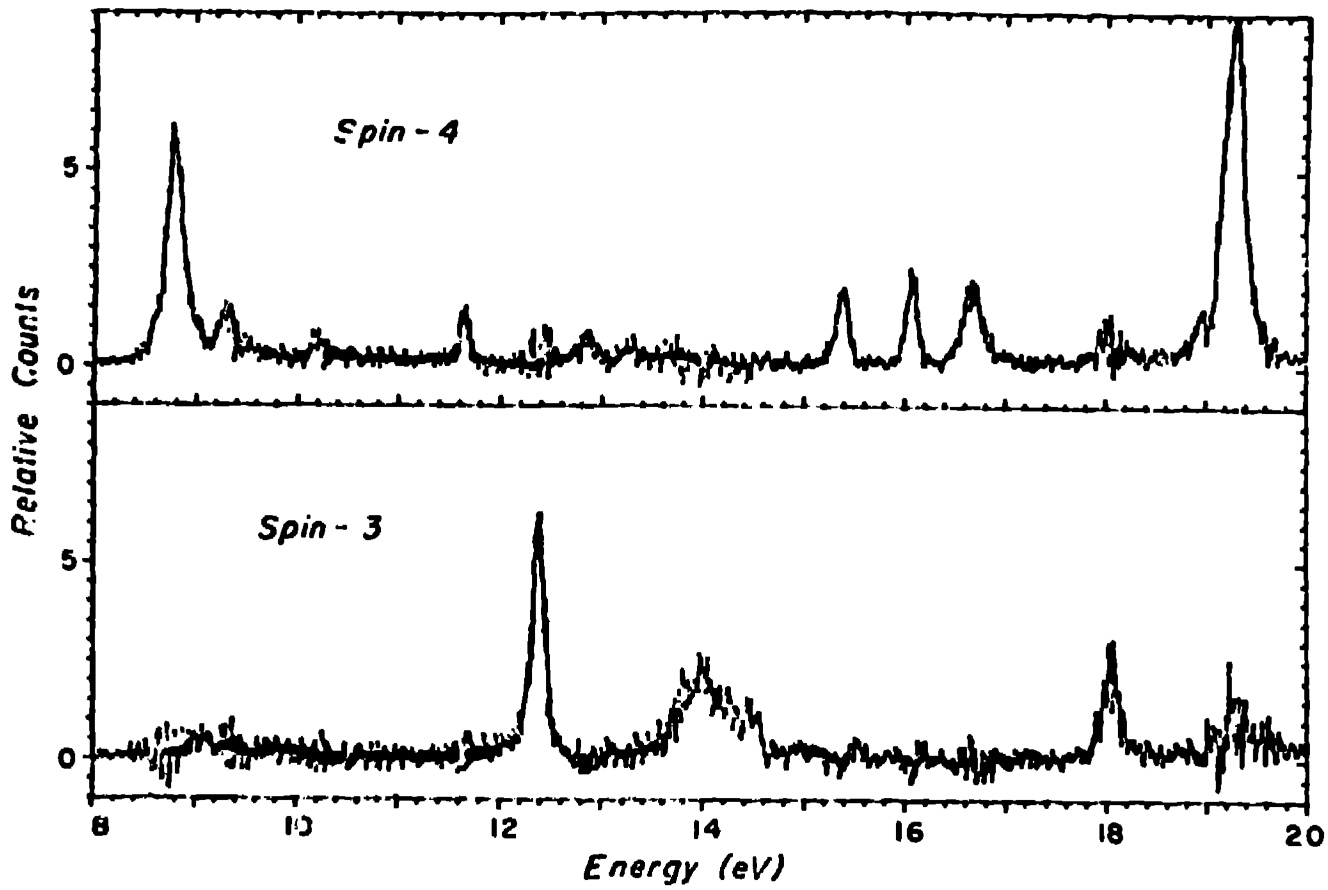

Fig. 1. Spin-separated resonance structure in the fission of $^{235}U + n$ versus neutron energy in the energy range from 8 to 20 eV. Note the presence of the very weak resonance with J = 3 at 9 eV. This resonance has not been seen previously due to the masking effect of the two resonances at 8.8 and 9.3 eV, each with J = 4.

In Fig. 5, we have plotted a stairstep distribution of spacings for resonances with J = 3 and J = 4 below 360 eV. The distributions have constant slope up to 60 eV, and the ratio of the slopes is close to what one expects if the level densities are proportional to (2J + 1). This suggests that few levels are missed below 60 eV. We applied the Δ_3 test of Dyson and Mehta[13] which also indicated that few levels are missed. By requiring that the Δ_3 statistic agree with the value predicted using the Gaussian Orthogonal Ensemble of Dyson,[14] we found probable positions for these few missing levels. With this technique, we arrived at the recommended average spacing of 1.153 eV and 0.896 eV for the J = 3 and J = 4 cases, respectively. This implies a total of 119 levels below 60 eV. As an independent check, we applied a missing level test which is based upon two assumptions:

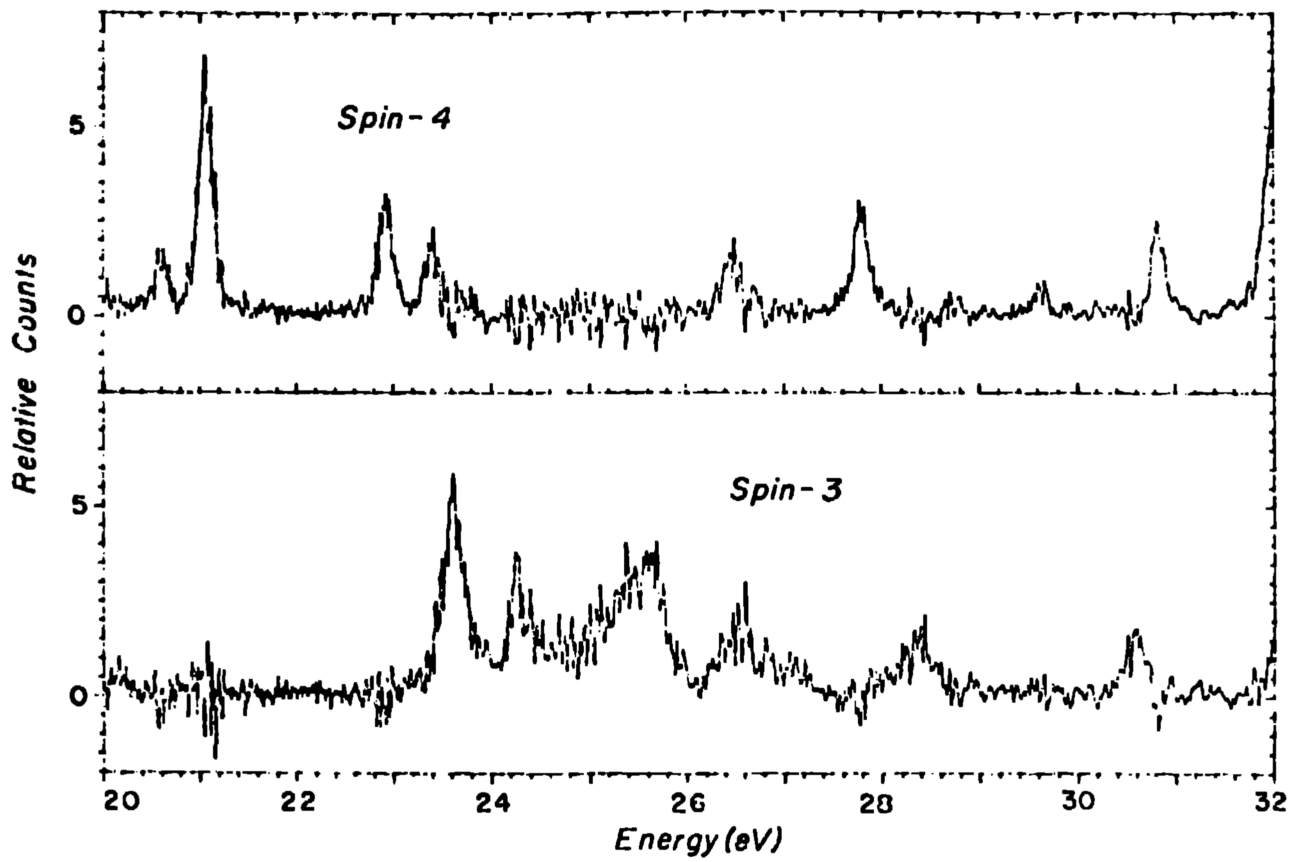

Fig. 2. Spin-separated resonance structure in the fission of $^{235}U + n$ versus neutron energy in the energy range from 20 to 32 eV. Note the doublet composed of a resonance with J = 4 at 26.4 eV and another with J = 3 at 26.55 eV. Although a comparison of the capture and fission cross sections does indicate two slightly displaced levels, this doublet has not been previously reported.

1) the neutron width distribution is Porter-Thomas, and 2) the widths larger than $\langle \Gamma_n^o \rangle /4$ are accurately known. With these assumptions, and the resonance parameters for ^{235}U of Reynolds,[15] we estimate that there are 110 ± 10 levels below 60 eV, in reasonable agreement with the estimate from the Δ_3 test. We thus feel confident that we have identified and assigned spins to a complete set of resonances in ^{235}U below 60 eV. The number of levels which are missed in the usual type of measurement, in which the spins are not separated, seems to be substantially lower than the statistical analysis of Garrison[16] would indicate. We also see no evidence for a very large number of missing levels as suggested by Felvinci et al.[17] For energies up to 350 eV we have assigned spins to most of the observed structure, although most individual resonances above 200 eV are unresolved.

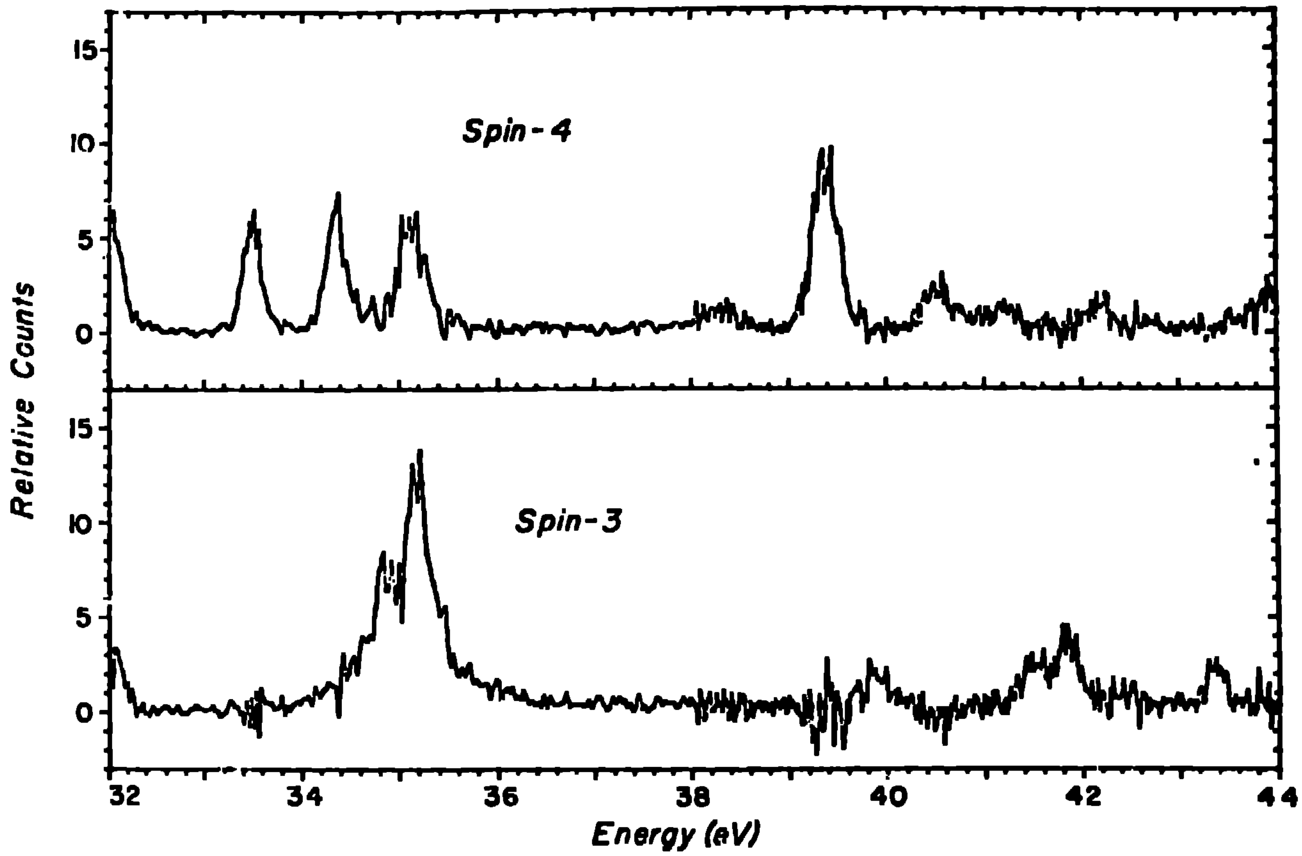

Fig. 3. Spin-separated resonance structure in the fission of $^{235}U + n$ versus neutron energy in the energy range from 32 to 44 eV. Note the separation of the complex structure in the vicinity of 35 eV.

Two sets of resonance parameters resulting from multilevel analysis of total and all measured partial cross sections are available: 1) those of Smith and Young[18] for ENDF/B-III, and 2) those of Reynolds for ENDF/B-V. Using the Smith and Young fission widths, we calculate $\langle \Gamma_f \rangle_{3-} = 0.179$ eV and $\langle \Gamma_f \rangle_{4-} = 0.090$ eV, whereas from Reynold's parameters we get $\langle \Gamma_f \rangle_{3-} = 0.220$ eV and $\langle \Gamma_f \rangle_{4-} = 0.098$ eV. This discrepancy can be attributed to the different values for the radiation widths of $\langle \Gamma_\gamma \rangle = 0.0355$ eV determined by Smith and Young and $\langle \Gamma_\gamma \rangle = 0.042$ eV assumed by Reynolds. The Bohr-Wheeler estimate, modified for a double-humped barrier, is expressed by

$$\langle \Gamma_f \rangle = \frac{n\langle D \rangle}{4\pi} \tag{4}$$

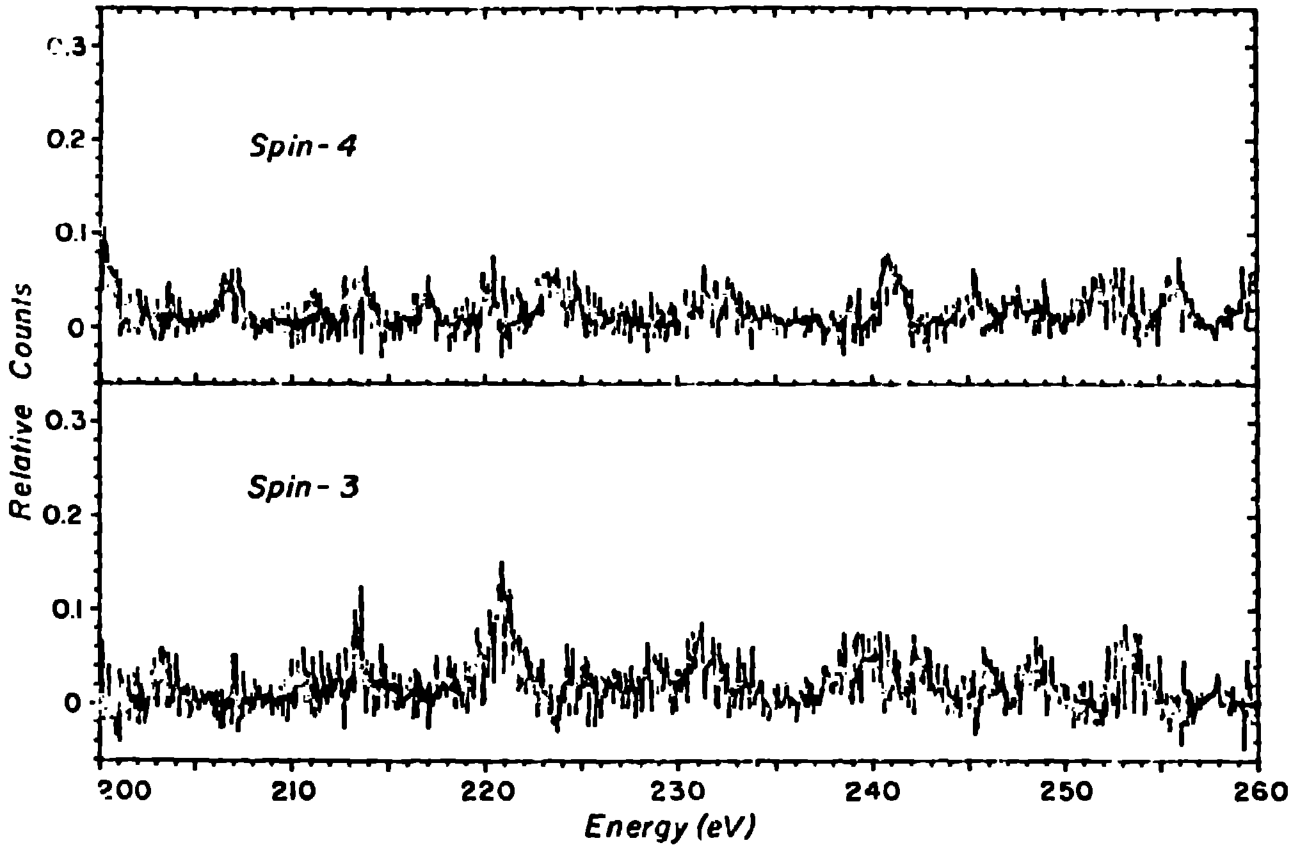

Fig. 4. Spin-separated structure in the fission of ^{235}U + n versus neutron energy in the energy range from 200 to 260 eV.

where n corresponds to the number of open channels. Using this expression, we arrive at fission widths of $\langle\Gamma_f\rangle_{3^-} = 0.092$ eV and $\langle\Gamma_f\rangle_{4^-} = 0.071$ eV for each open channel. The results of the two multilevel analyses are consistent with approximately two open channels for J = 3, or more if the channels are only partially open, and with no more than one fully open channel for J = 4 resonances.

Additional information regarding the configuration of those fission channels may be gleaned from the Pattenden and Postma[10] data on the angular distribution of fission fragments from aligned ^{235}U. This angular distribution may be expressed as

$$W(\theta) = 1 + \sum_{\substack{n \text{ even} \\ n \leq 21}} A_n f_n(I) P_n(\cos \theta) \qquad (5)$$

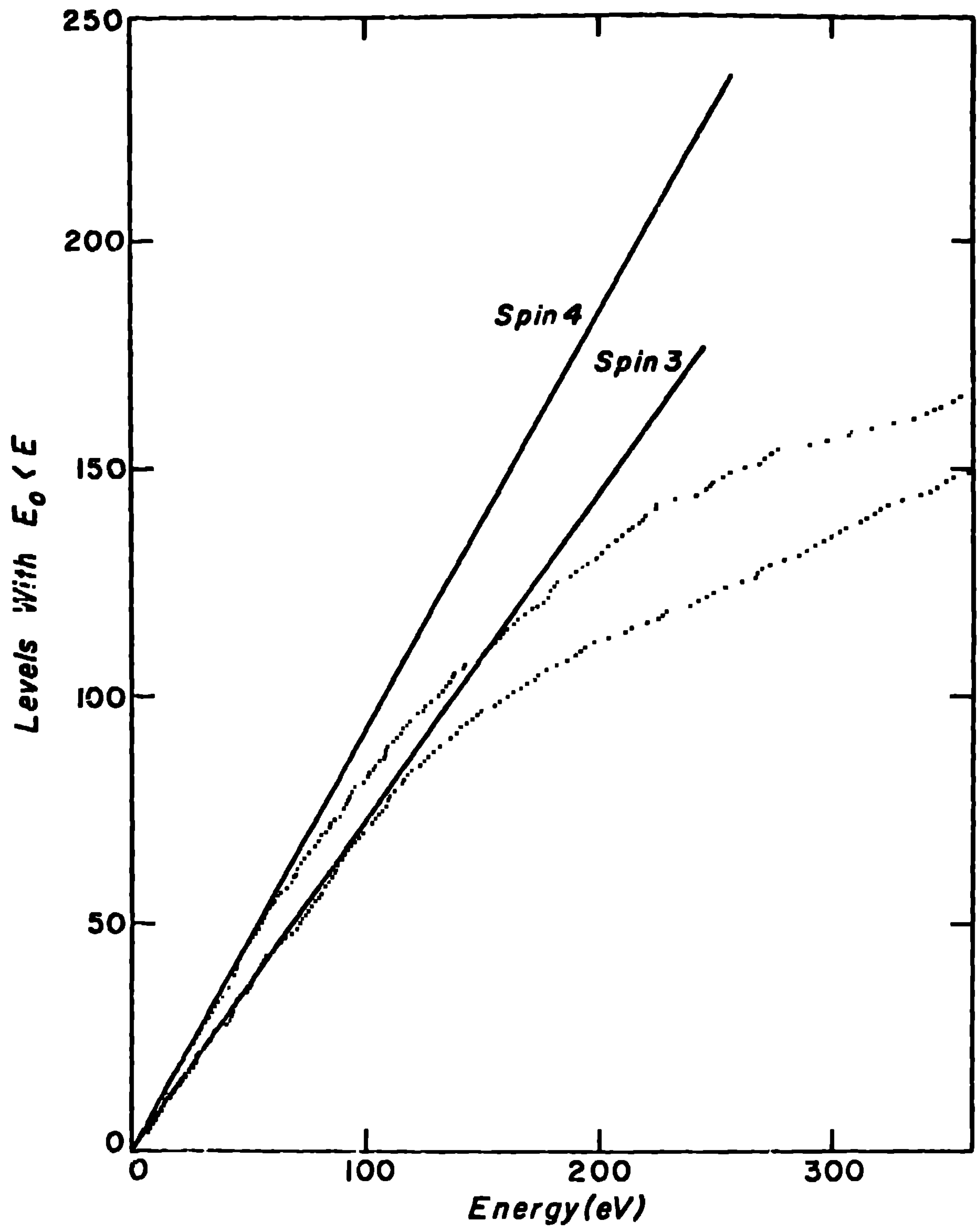

Fig. 5. Observed resonance spacing distribution in (^{235}U + n) below 360 eV. Data points give the number of levels having a resonance energy less than the energy shown on the abscissa, and correspond to the tips of the stairs in the usual stairstep plot. The solid lines represent a fit to the data points below 60 eV, and show the expected (2J + 1) slope.

Where the A_n parameters contain the geometric factors, the f_n are the alignment parameters, and P_n are the Legendre polynomials. In the Pattenden and Postma measurements, only the A_2 terms are measured. For a well-resolved resonance with known spin and unique K, measurement of A_2 should determine K. Because of the use of a fission booster target with a relatively long pulse of 230 ns and a short (10 m) flight path, few of the resonances in the Pattenden and Postma experiment were well resolved. Thus, we define an average or effective J value for $^{235}U + n$ as

$$J_{eff} = 3 + \frac{\sigma_4}{\sigma_3 + \sigma_4} \tag{6}$$

where σ_3 and σ_4 are the spin-3 and spin-4 cross sections used in Eqs. (2a) and (2b). A plot of A_2 versus J_{eff} is shown in Fig. 6. The solid line in the figure represents a least-squares fit to the data and may be used to infer the average value of A_2 for pure spin-3 resonances (J_{eff} = 3.0) and for pure spin-4 resonances (J_{eff} = 4.0). We thus obtain $\langle A_2 \rangle_{J=3}$ = 1.22 and $\langle A_2 \rangle_{J=4}$ = 2.01. Knowing that the (J,K) = (4,0) channel is forbidden because of parity conservation and recalling the assumptions from above on the number of open channels for each spin state, we may assume that the two lowest channels, (J,K) = (4,1) and (4,2), are open. Knowing the A_2 value for each (J,K), we may calculate the contribution from each channel. If for the average fission widths we take the mean of the averages from the two multilevel analyses, we get $\langle \Gamma_f \rangle_{J=3}$ = 0.20 eV and $\langle \Gamma_f \rangle_{J=4}$ = 0.094 eV. For the J=4 resonances, we determine $\langle \Gamma_f \rangle_{J,K=4,1}$ = 0.075 eV and $\langle \Gamma_f \rangle_{4,2}$ = 0.019 eV. This implies that since the (J,K) = (4,1) channel is fully open, the (3,1) channel should also be fully open with a fission width of .096 eV. Solving for the K = 0 and 2 channel widths, we get $\langle \Gamma_f \rangle_{J,K=3,0}$ = 0.020 eV and $\langle \Gamma_f \rangle_{3,2}$ = 0.084 eV. However, the assumption of $\langle \Gamma_f \rangle_{3,0}$ = 0 and thus $\langle \Gamma_f \rangle_{3,1}$ = $\langle \Gamma_f \rangle_{3,2}$ = 0.100 eV is not inconsistent with the errors involved. The surprising fact is that, although it has long been assumed that the channels open in order of ascending K, following the sequence of octupole bands observed near the ground states of even-even nuclei. Why the (J,K) = (3,0) channel is either completely or nearly completely closed can presently only be answered hypothetically.

Although we know that the average behavior is consistent with the above explanation based on the fission channel concept, we do not yet know whether K is a conserved quantum number in fission. One notes in Fig. 6 that the points are nearly uniformly distributed over a broad range of A_2 values. This implies that the observed angular distribution is not consistent with integer K-values, but rather there is an admixture of the fission channels. However, one must be wary of overinterpreting the Pattenden and Postma results due to the lack of well-resolved resonances in this data.

The preceding discussion addresses only the resolved region in ^{235}U. It has been suggested [20,21] that the fluctuations in the unresolved region result from local enhancement due to broad states in the second well of the double-humped fission barrier. Keyworth et al[4] showed that for $^{237}Np + n$, the subthreshold fission resonances corresponding to a single state in the second well all have the same spin. If the structure in $^{235}U + n$ involves a similar mechanism, then one would expect a spin dependence.

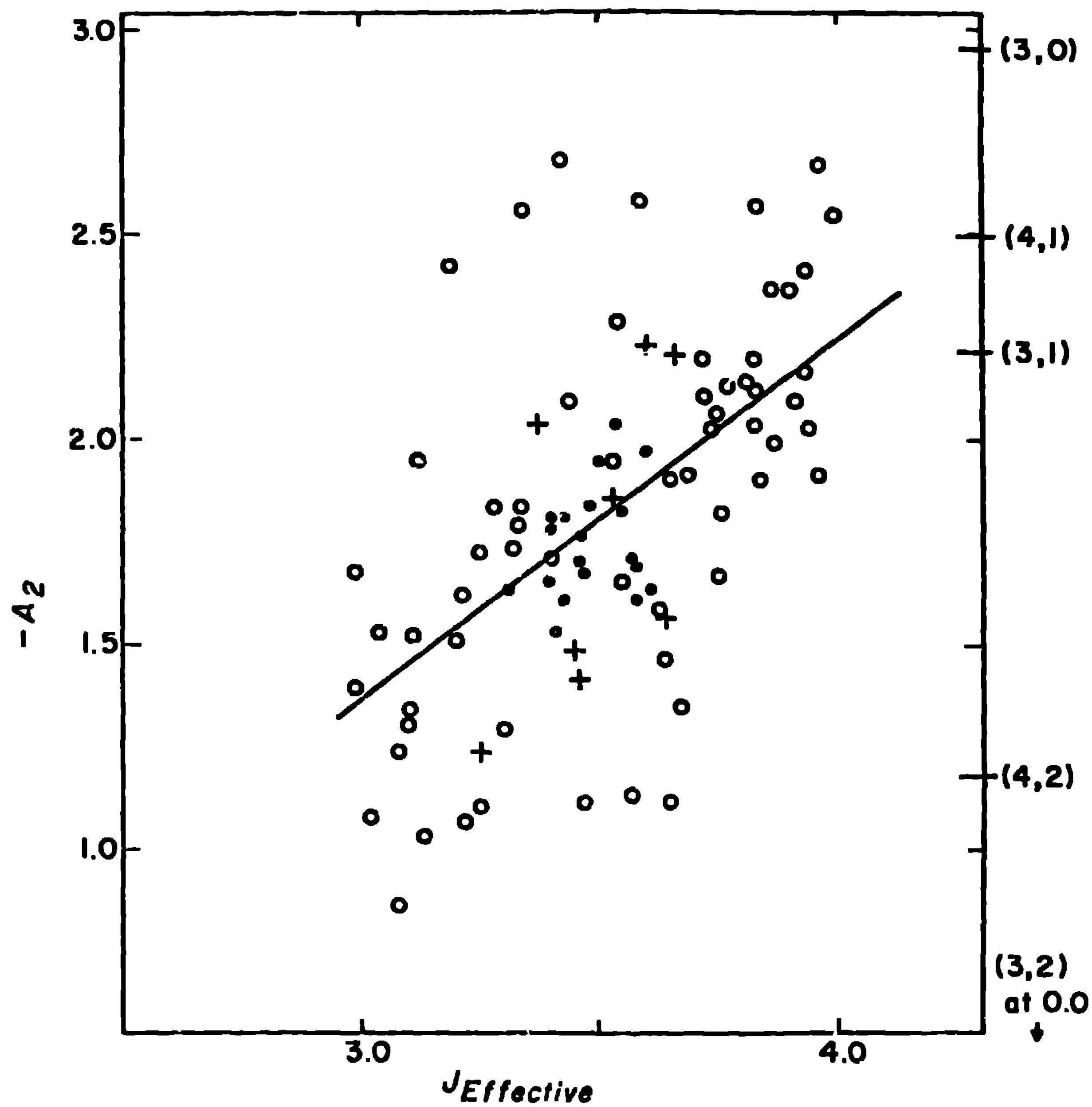

Fig. 6. The variation of A_2 from Pattenden and Postma versus $J_{effective}$ = $3 + \sigma_4/(\sigma_3 + \sigma_4)$. The straight line shows a linear least-squares fit to these data. The open circles show A_2 data for resonance structure, the closed circles data for the unresolved region below 2 keV, and the plus signs data for the between-resonance background regions reported by Pattenden and Postma.

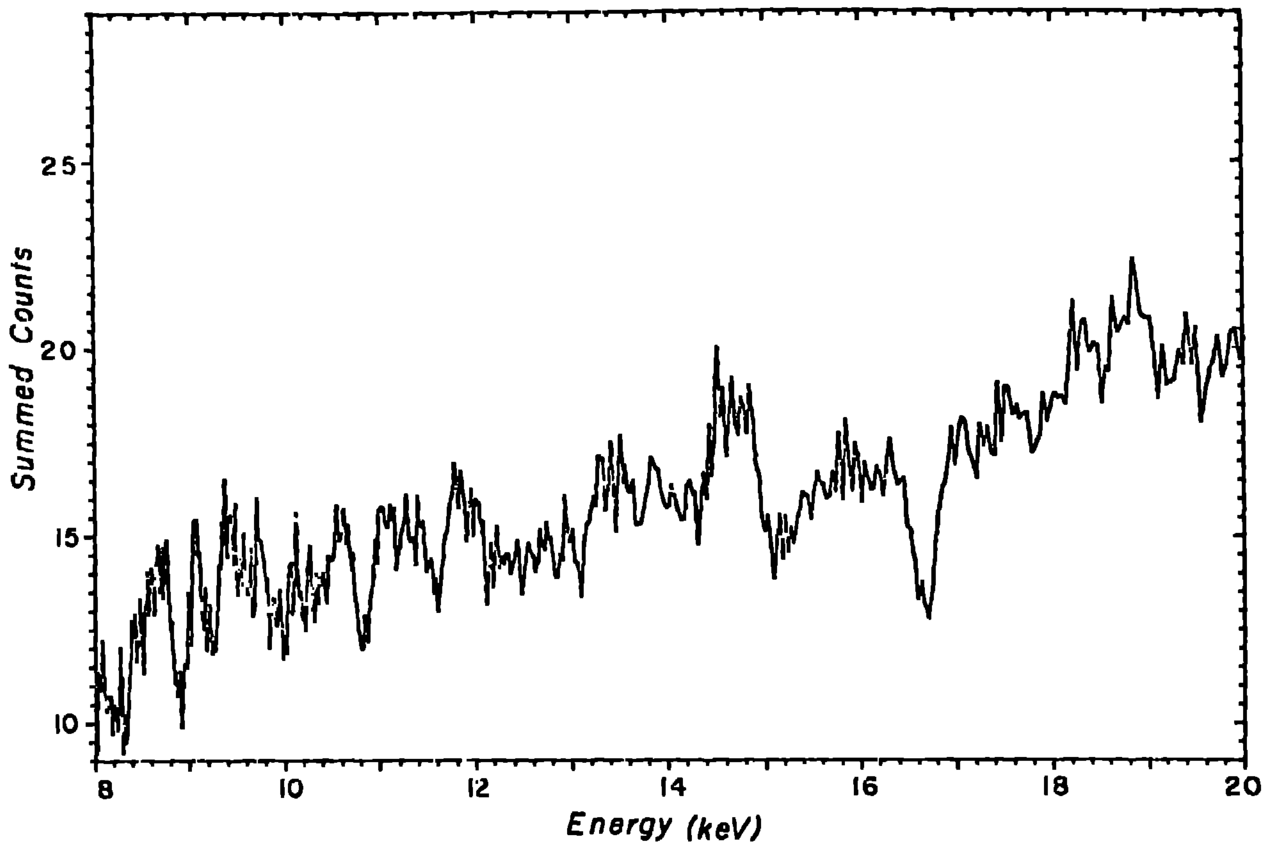

Fig. 7. Summed counts (spin-3 enhanced plus spin-4 enhanced count rates) observed in the fission of (^{235}U + n) versus neutron energy in the energy range from 8 to 20 eV.

The fission cross section of ^{235}U + n in the range 8 - 20 keV is shown in the plot of summed counts, $N_3 + N_4$, in Fig. 7. The large fluctuations are clearly evident. However, the spin-separated data over the same energy region, shown in Fig. 8, show minimal evidence for any spin dependence in the fluctuation, possibly due to the poor statistical accuracy. To test quantitatively for intermediate structure, we then pursued statistical tests on broad-bin averages. Following Migneco et al,[20] we initially carried out a Wald-Wolfowitz runs-distribution test from 0.1 to 25 keV on $J_{eff} - \langle J_{eff} \rangle$ using bins of 240 and 400 eV, and from 0.1 to 10 keV with bins of 85 eV. Although Migneco et al reported that this test gave significant results when applied to σ_f for ^{235}U, the test applied to the polarization data gave results consistent with a random distribution of spin. A similar calculation of the serial correlation coefficients of J_{eff} followed by a Wald-Wolfowitz test on these coefficients again showed no significant departure from a random distribution.

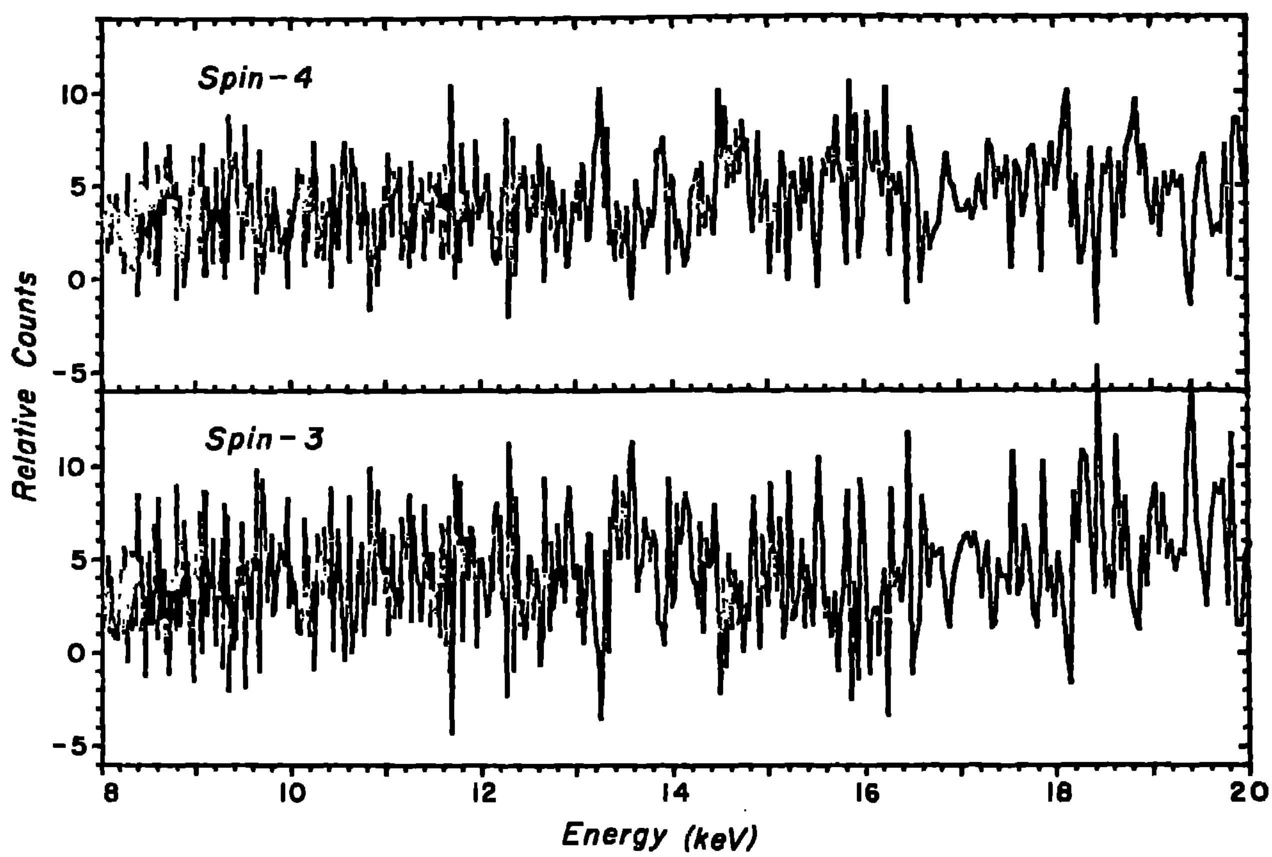

Fig. 8. Spin-separated count rates in the fission of (^{235}U + n) versus neutron energy in the energy range from 8 to 20 keV. Except for the cluster between 14 and 15 keV, which is clearly spin 4, it is not obvious that either of these curves correlates with that shown in Fig. 7.

Another test, however, showed a more interesting result. We calculated the correlation coefficient between the spin-3 data and the summed counts and between the spin-4 data and the summed counts, for broad-bin averages. The results, shown in Table I, imply that the observed structure is attributable to spin 4. Apparently, there is still enough statistical error associated with the broad-bin averages that it masked the effect when we used the usual tests for intermediate structure. We do feel, however, that the results shown in Table I are definitive and show that essentially all the fluctuating part of the ^{235}U fission cross section in the region analyzed has J=4. We thus conclude that these polarization data give strong support to the hypothesis that the fluctuations in the ^{235}U fission cross section are a second-well phenomenon.

CONCLUSIONS

Knowledge of the $^{235}U + n$ system has been substantially enhanced by removing uncertainties in the resonance spins. For an understanding of the average properties, division of the resonances into the appropriate spin groups permits an accurate description of the cross section. However, understanding of the underlying fission process remains incomplete.

For example, with this spin information, one may search for a dependence upon J and K of the fission fragment mass distribution, fragment kinetic energy distribution, and $\bar{\nu}$. Although available measurements of these quantities are limited in resolution and scope, there is no clear evidence for dependence upon spin alone. In fact, measurements[21,22] of $\bar{\nu}$ over several of the larger resonances in ^{235}U clearly preclude a dependence of this quantity upon J alone. However, it has been demonstrated[23] that the (n,γf) process can account for the relatively large fluctuations of $\bar{\nu}$ in ^{239}Pu. Although the evidence[24] is less persuasive in the case of ^{235}U, this process is probably involved in the $\bar{\nu}$ variations. One may well expect that the fragment mass and kinetic energy distributions are dependent upon J and K but the poor state of knowledge of K values coupled with the poor resolution in existing measurements makes detailed interpretation tenuous.

What is needed is a coherent approach toward answering these questions, initially in ^{235}U alone. Using the new time-of-flight facility being implemented at the Los Alamos Meson Physics Facility (LAMPF), the Weapons Neutron Research Facility (WNR), we are pursuing such an approach. Using an antiferromagnetic intermetallic compound of uranium rather than the paramagnetic crystals used by Dabbs and Pattenden and Postma, we hope to achieve sufficient alignment of ^{235}U with sufficient resolution to determine the K-value, or the admixture of K-values, for each J-value assigned in the measurements using a polarized target and beam. Concurrently, we will use the intense low energy neutron flux at the WNR facility to determine the fragment kinetic energy distribution and the fragment mass distribution for the larger resonances in ^{235}U. We expect that the results of these proposed measurements, in conjunction with existing data, will provide answers to those questions on ^{235}U addressed earlier in this paper.

TABLE I

Correlation coefficients and significance levels for the correlation of spin-3 and spin-4 data with structure in ^{235}U σ_f, from 8 - 25 keV. In this table, the significance level is the probability that the observed correlation or larger would occur with a randomly selected sample.

Energy Range (keV)	Bin Width (keV)	$\rho(N_3,\Sigma)$	Significance of $\rho(N_3,\Sigma)$	$\rho(N_4,\Sigma)$	Significance of $\rho(N_4,\Sigma)$
8.0 - 10.4	0.12	-0.01617	∿0.50	0.7048	0.0003
10.4 - 12.8	0.12	0.2148	0.18	0.6148	0.002
12.8 - 15.2	0.12	0.0889	0.35	0.3815	0.05
15.2 - 20.0	0.24	0.1996	0.20	0.7111	0.0002
20.0 - 24.8	0.24	0.2336	0.16	0.7443	0.0001
24.8 - 34.4	0.48	0.2864	0.11	0.8194	<0.00001

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