$I=2^-$. Therefore, the gamma-ray transition to the $K=0$, $I=2^+$, $E=0.123$-MeV state could be electric dipole plus magnetic quadrupole.

Now, the dependence of the anisotropy coefficient on the multipolarity admixture δ is not single valued. The experimental result of $A=-0.21\pm0.07$ is consistent with a value of $\delta>1$ or with a value of $\delta\ll1$. Baba and Bhattacherjee were able to establish the $E1$ character of the 1.28-MeV gamma transition. Hence, the large value of δ can be excluded.

The assignment $K=0$ to the 1.4-MeV state can be eliminated due to symmetry conditions. It is further noted that the assignment $K=2$ to the 1.4-MeV state would leave the electric dipole K forbidden while the magnetic quadrupole is allowed. The result is that the $E1$ transition would be retarded which would permit a large admixture of $M2$ into the transition. This has been observed in a similar gamma transition of the 1189-keV level in W^{182} with a 40% $M2$ admixture.[9]

However, the assignment of $K=1$ to the 1.4-MeV level would greatly reduce the $M2$ admixture with the result that $M2/E1$ should be less than unity by several orders of magnitude.

[9] C. J. Gallagher and J. O. Rasmussen, Phys. Rev. **112**, 1730 (1958).

We note the experimental $M2/E1$ admixture gives the $M2$ intensity as 2% or less and consistent with zero which implies the K value is $K=1$. In addition the abnormally large $\log ft=10.1$ could be explained by a $K=1$ assignment to the 1.4-MeV level. Since the ground state of Eu^{154} is a $K=3$ state, the beta decay to the 1.4-MeV level would be a second-order K forbidden transition and thus would be greatly retarded. Also, the $K=2$ assignment is not indicated by the ratio of the ft values for the beta transitions to the 1.72-MeV level and the 1.4-MeV level. A $K=2$ assignment to 1.4-MeV level would give this ratio a value of around one while the experimentally observed ratio is ~0.07. On the basis of the above discussions a tentative assignment of $K=1$ is given to the 1.4-MeV level.

ACKNOWLEDGMENTS

The authors wish to express their sincere gratitude to Dr. Lawrence Gallaher for suggesting this problem and for his continued guidance and interest in this work.

The authors are endebted to Oak Ridge National Laboratory for the service irradiation, and to James Howes, Battelle Memorial Institute, Columbus, Ohio, who performed the chemistry necessary for the source deposition.

PHYSICAL REVIEW VOLUME 134, NUMBER 5B 8 JUNE 1964

Fluctuations in the Partial Radiation Widths of U^{239}†

H. E. Jackson
Argonne National Laboratory, Argonne, Illinois

(Received 29 January 1964)

The Argonne fast chopper has been used to measure the distribution of the sum of the intensities of transitions having energies between 3.8 and 4.2 MeV in the neutron-capture spectra of 12 neutron resonances of U^{238}. The pulse-height spectra were observed in a large (8×6 in.) NaI(Tl) scintillator. The partial radiation widths exhibit large fluctuations. If the observed distribution is characterized by a number ν of degrees of freedom, as in the theoretical treatment by Porter and Thomas, a value of $\nu=5.8\pm2.3$ is obtained. This is in contrast to the previously reported results which range from 11 to 90 and is consistent with the predictions of the simple statistical model for the distribution of the sum of four uncorrelated partial radiation widths.

I. INTRODUCTION

THE study of the statistical properties of partial radiation widths for nuclei excited by neutron capture has been of central interest in slow-neutron spectroscopy for the last four years. From the various experiments[1–4] on a wide range of nuclei, with one exception clear and consistent conclusions have been obtained. The distribution that governs the partial radiation widths for transitions from resonances of a given spin and parity to a definite final state is characterized by large fluctuations about a mean value; widths with values less than the mean predominate. It has been customary to analyze experimental data in terms of the family of χ^2 distributions with ν degrees of freedom, as proposed by Porter and Thomas.[5] The Porter-Thomas distribution, which is in good agreement with data on reduced neutron widths corresponds

† Work performed under the auspices of the U. S. Atomic Energy Commission.

[1] J. Julien, C. Corge, V. D. Huynh, F. Netter, and J. Simic, J. Phys. Radium **21**, 423 (1960).

[2] F. D. Brooks and J. R. Bird, in *Proceedings of the RPI Neutron Physics Symposium, Troy, New York, 1961*, edited by M. L. Yeater (Academic Press Inc., New York, 1962), p. 109.

[3] R. E. Chrien, H. H. Bolotin, and H. Palevsky, Phys. Rev. **127**, 1680 (1962).

[4] L. M. Bollinger, R. E. Coté, R. T. Carpenter, and J. P. Marion, Phys. Rev. **132**, 1640 (1963).

[5] C. E. Porter and R. G. Thomas, Phys. Rev. **104**, 483 (1956).

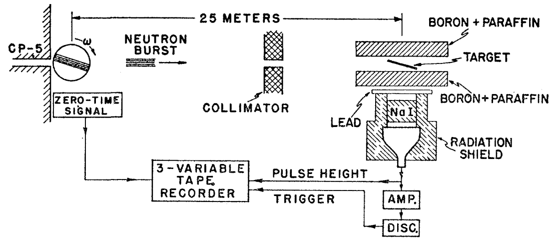

FIG. 1. Schematic diagram of the experimental arrangement.

to $\nu=1$; the distribution function for $\nu=2$ is a simple exponential. For most nuclides, the various experimenters are in general agreement that the most probable value of ν is between 1 and 2. This result is not unexpected since the radiative transition to a definite final state is usually assumed to be a single-channel process. In the statistical model of compound states, the Porter–Thomas distribution will describe the behavior of the partial widths for a single-channel process. Therefore, we expect $\nu=1$ for the distribution of partial radiation widths as well as that of reduced neutron widths.

However, one striking anomaly has remained, namely U^{238}. This nuclide, whose ground state has $J=0$, was a particularly convenient case because all resonances excited by s-wave neutron capture have the same spin, $J=\frac{1}{2}$. In terms of its reduced width, the 4.06-MeV transition observed in the thermal capture of U^{238} is one of the strongest $E1$ transitions known.[6] Thus, it was natural to attempt a study of the distribution of the partial widths for this transition for a large number of resonances. Such studies, which were carried out by groups at Brookhaven and Saclay,[7,8] indicated fluctuations of less than about 11% about the mean—in strong contrast to the results for all other nuclei. These results imply a value of ν between 50 and 100. A more recent unpublished analysis[9] of the same Brookhaven data indicates $\nu=11$, which is still anomalously high. Two later experiments were performed to estimate the number of excited levels of U^{239} populated by primary transitions of about 4.0 MeV. Detailed examination of resonance spectra by the Argonne group[10] and coincidence studies of the thermal spectrum by a Brookhaven group[11] indicate four transitions that contribute to the 4.06-MeV peak. However, even these facts do not explain the reported uniformity of the uranium gamma rays, since the data imply the presence of at least 11 lines near 4.0 MeV if their individual widths are uncorrelated.

However, the case of U^{239} differs from the other nuclides studied in one essential respect: the capture spectra for U^{239}, as indicated by the level scheme of Fiebiger,[11] are much more complex than those of any of the other nuclei. At best, the techniques available to the earlier experimenters were marginally adequate for such a case. Since that time two experimental advances have been made, namely large-volume scintillators with improved response functions, and improved methods of analysis of data. Even now the techniques are not sensitive enough to permit a determination of the individual partial widths from single-photon resonance-capture spectra. However, the sum of the partial widths previously measured as a single transition can be measured with sufficient precision. For these reasons, an attempt (reported here) was made to confirm the anomaly in U^{239} in an independent measurement. In contrast to earlier experiments, we observed large fluctuations in the measured intensities, and the data are consistent with the results reported for other nuclei.

II. APPARATUS

The experimental arrangement is shown schematically in Fig. 1. The Argonne fast chopper[12] was used to select neutrons of known energy by measurement of their time of flight. The sample consisted of six 20-mil plates of highly depleted uranium, placed at a large angle with respect to the neutron beam in order to be effectively thick to the incoming neutron beam and thin for capture photons. The sample was placed 25 m from the chopper. The duration of the neutron burst from the chopper was about 1.6 μsec, and the over-all resolution of the system was about 0.08 μsec/m for neutron energies above 100 eV. Capture γ rays were observed in a shielded NaI(Tl) crystal 8 in. in diameter and 6 in. deep which was placed 8 in. from the center of the sample. The beam was collimated in such a manner that the irradiated portion of the sample was about the same size as the face of the crystal. To minimize background caused by neutrons scattered into the crystal, the sample was mounted in the center of a rectangular tube with 3-in.-thick walls of a mixture of boron carbide and paraffin. In addition, a $\frac{1}{4}$-in. plate of lead was placed over the face of the crystal to attenuate the high background caused by the natural activity of the uranium sample. This thickness was chosen to produce the maximum attenuation of the lower energy background (E_γ<500 keV) consistent with no serious deterioration of the response function of the NaI(Tl) crystal for photons of energies between 4 and 5 MeV.

---

[6] G. A. Bartholomew, Ann. Rev. Nucl. Sci. **11**, 274 (1961).

[7] D. J. Hughes, H. Palevsky, H. H. Bolotin, and R. E. Chrien, in *Proceedings of the International Conference on Nuclear Structure, Kingston, Canada, 1960*, edited by D. A. Bromley and E. W. Vogt (University of Toronto Press, Toronto, 1960), p. 771.

[8] C. Corge, V. D. Huynh, J. Julien, J. Morgenstern, and F. Netter, J. Phys. Radium **22**, 722 (1961).

[9] R. E. Chrien, in Proceedings of the International Conference on Nuclear Physics with Reactor Neutrons, Argonne National Laboratory Report ANL-6797, 1963, p. 339 (unpublished).

[10] H. E. Jackson and L. M. Bollinger, Bull. Am. Phys. Soc. **6**, 274 (1961).

[11] N. F. Fiebiger, Institute für Kernphysik Frankfurt/M Report IKF-8, 1963 (unpublished).

[12] L. M. Bollinger, R. E. Coté, and G. E. Thomas, Proc. 2nd Intern. Conf. Peaceful Uses At. Energy, U. N., Geneva **14**, 239 (1958).

The pulse-height gain of the system was continuously monitored by electronic scanning of the 662-keV peak produced by a Cs^{137} source which was placed on the face of the crystal. Any drift in gain of the system was automatically compensated by applying a correction to the high voltage on the phototube. With the exception of the 6.7-eV resonance, all resonance spectra were recorded simultaneously in a single run under identical conditions of pulse-height gain and time-of-flight calibration. This was accomplished by use of the Argonne 3-variable magnetic-tape recording system which has been described in detail by Rockwood and Strauss.[13] Each event, signaled by the arrival of a discriminator pulse at the recording section of the analyzer, was recorded on a magnetic tape in the form of a 9-bit binary number representing the time of flight and an 8-bit number representing the pulse height. All events with pulse heights corresponding to energies greater than 3.0 MeV and every fourth event with an energy between 1.8 and 3.0 MeV were recorded. Because the only information of interest in the low-energy region is the total number of counts, the limited storage capacity of the magnetic tape was used most efficiently by this mode of selection of events.

The record on the tape was analyzed on the search unit of the magnetic-tape system. The data were scanned and the spectra of one variable were obtained for arbitrary conditions on the values of the other variable. As an example, Fig. 2 shows the time-of-flight spectrum as obtained from the search section when the γ-ray pulse height is restricted to energies between 3.0 and 4.8 MeV. To obtain the resonant-capture spectra, the time of flight was restricted to values corresponding to locations of the various resonances observed in the time-of-flight spectrum. The background for each resonance was obtained by interpolation between the spectra recorded for neighboring time-of-flight regions that contain no resonance structure.

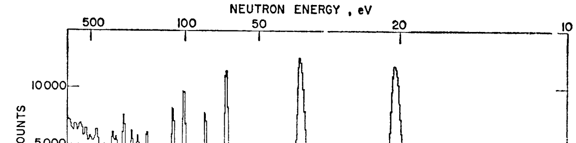

FIG. 2. Time-of-flight spectrum for events with γ-ray pulse heights between 3.0 and 4.8 MeV. In addition to the resonances of interest, one at 10.2 eV (usually assumed to be excited by p-wave neutrons) can be seen. However, the small number of events prevented detailed observation of the capture spectrum.

The spectrum for the 6.7-eV resonance was obtained from a separate run that covered neutron energies less than 120 eV. The spectra for resonances at 21, 37, 66, and 103 eV were used to normalize the results to those for the run shown in Fig. 2.

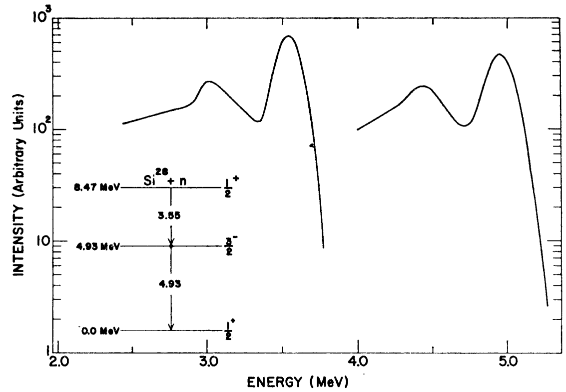

FIG. 3. Line shapes for 3.55- and 4.93-MeV photons. Shown at the lower left is the level diagram of the gamma-ray cascade following neutron capture in Si^{28}; it was used to obtain the line shapes.

III. EXPERIMENTAL PROBLEMS

Two subsequent measurements were required before the observed pulse-height spectra could be analyzed to obtain relative partial widths: (1) an accurate measurement of the response function of the NaI crystal to monenergetic gamma rays, and (2) an estimate of the component of the single-photon spectrum which results from summing events, i.e., individual cascades in which two photons are observed as a single photon.

The response function should be obtained for a sufficient number of photon energies to ensure an accurate estimate of the line shape at the energies of transition observed in the resonance spectra of uranium. In the energy range of interest, 3.5–5.0 MeV, the changes in the line shape are gradual and can be determined with sufficient precision by a linear interpolation of the shapes measured for two energies that bound the range of energies involved in the analysis of the uranium data. These two shapes were obtained from measurement of the thermal-capture spectrum of Si^{28}, which is dominated by a two-step cascade through the 4.93-MeV state directly to the ground state of Si^{29}. The capture spectrum of a SiO_2 sample was observed in two identical large NaI crystals operated in coincidence. The data were recorded and analyzed by means of the magnetic-tape system. Line shapes for 3.55- and 4.93-MeV photons (Fig. 3) were obtained by restricting the pulse heights in the second crystal to the region of the photopeak of the accompanying member of the cascade.

The summing component in the spectra was estimated in two experiments. In one measurement, the capture spectrum of the 6.7-eV resonance was measured with a solid angle which was 0.25 of that used in the complete measurement. The resulting spectrum was not significantly different from the one obtained with the larger solid angle.

The summing contribution was also measured directly for each resonance studied by a sum-coincidence experiment. The pulse heights observed in two identical

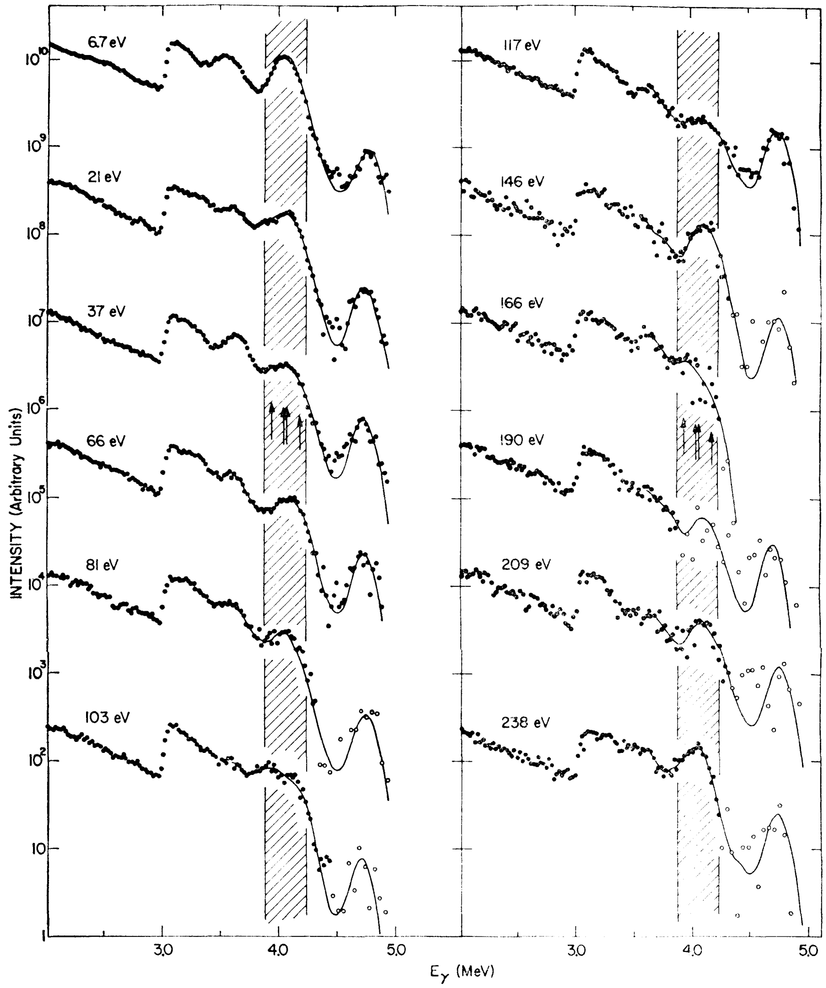

FIG. 4. Pulse-height spectra for resonances in U^{238}. The open circles represent data points obtained by averaging adjacent channels. The solid curves through the data points are the least-squares fits made to obtain the intensities of the individual transitions.

NaI crystals were added to obtain a direct measure of the sum spectra for each resonance. The results indicated that no correction of the data for summing was necessary. In the energy range of primary interest, 3.0 to 4.3 MeV, the summing component in the experimental single-phonon spectra was found to be small compared with the statistical errors in the data. Typically, such events represented less than 2% of the counts per channel. In the neighborhood of the neutron binding energy of U^{238}, 4.60 to 4.80 MeV, the summing component was on the average 10% of the observed single rate. Subtraction of these events did not appear justified in view of the poor statistical quality of the data in this region. The weak intensity of summing events may be explained in part by the fact that the amount of absorbing material placed between the crystal and target was chosen to cut out all radiation below about 500 keV. Consequently, summing resulting from simultaneous observation of low-energy photons and relatively strong transitions with energies between 3.0 and 4.2 MeV was inhibited.

IV. RESULTS AND ANALYSIS

A compilation of the capture spectra with background subtracted is shown for twelve resonances in Fig. 4. The shaded area represents the region of interest in previous experiments. Two qualitative features are apparent immediately from inspection of these spectra. First, the abnormal strength of the transition at 4.06 MeV observed in the spectrum for the 6.7-eV resonance is not usually characteristic of the spectra for resonances. The observation of this transition in thermal capture that is dominated by this 6.7-eV resonance is what caused the early interest in the resonance-capture spectra. Second, the spectra include radiation above the 4.06-MeV transition; and this plus the effects of transitions at lower energies (for example, at 3.66 and 3.81 MeV) must be included in an accurate analysis of the shaded region. For this reason, the spectra were studied over the region from 3.5 to 5.0 MeV, and the effects of all known transitions in this range were included in the measurements of the partial widths of interest.

The intensities of the individual primary transitions were determined by a simultaneous least-squares fitting of the resonance spectra by use of a single set of transition energies and one set of response functions for the NaI crystal. The method of analysis has been described in detail in Ref. 4. The resulting intensities were then normalized to the same number of captures under the assumption that the observed number of counts in each spectrum in a low-energy region between 1.8 and 3.0 MeV is proportional to the capture rate for that resonance.

The energies of the primary transitions used in the analysis are shown in Fig. 5. All but the 4.74-MeV transition have been observed in thermal capture by Fiebiger.[11] Although the 4.74-MeV transition has not been reported before, the intensity of the peak in the various spectra is typically about 10 times the measured summing component, and it appears reasonable to interpret it as a primary transition to one or more levels near the ground state. Such a collective band of levels has been proposed by Emery *et al.*[14] although a slight discrepancy (70 keV) exists between the energies proposed by them and our observed energy. However, the neutron binding energy of U^{238} is known only to about this precision. [*Note added in proof*. This interpretation is confirmed by recent $U^{238}(d,p)U^{239}$ data reported by B. Macefield and R. Middleton, Argonne National Laboratory Report ANL-6848, 1964, p. 52 (unpublished). Their measured ground-state Q value gives a neutron separation energy of 4.81 ± 0.02 MeV for U^{239}. Measurements of the proton spectra show a band of levels in U^{239} at an excitation energy of about 100 keV.]

TABLE I. Results of analysis of capture gamma spectra for resonances in U^{238}. The measured number I (4.00 MeV) represents the relative value of the sum of partial widths for transitions in the energy range between 3.90 and 4.25 MeV; while I (4.70 MeV) represents the relative value of the sum of partial widths for transitions with energies greater than 4.60 MeV.

Resonance energy	I (4.00 MeV)	I (4.70 MeV)
6.7	2.14 ± 0.21	1.16 ± 0.12
22.0	1.37 ± 0.14	1.06 ± 0.11
37	0.84 ± 0.09	1.06 ± 0.12
66	0.71 ± 0.07	0.89 ± 0.11
81	0.62 ± 0.07	0.44 ± 0.12
103	1.08 ± 0.11	0.45 ± 0.12
117	0.41 ± 0.05	2.02 ± 0.20
146	1.04 ± 0.12	0.51 ± 0.34
166	0.78 ± 0.09	0.00 ± 0.20
190	0.28 ± 0.05	1.05 ± 0.30
209	0.68 ± 0.09	1.34 ± 0.30
238	2.02 ± 0.20	2.01 ± 0.31

For our crystal, the full width at half-maximum of the photopeak of the line at 4.0 MeV is 170 keV, much larger than the spacing of states of U^{239} which are fed by transitions with energies of about 4.0 MeV. Nevertheless, in the spectra of Fig. 4 it is apparent from the shift in the position of the peak near 4.0 MeV from resonance to resonance that a group of transitions similar to those observed by Fiebiger is present. Fluctuations in the individual partial widths would account for the shifts in the position of the composite peak. Because the spacing of the states being populated is small relative to the γ-ray resolution width, the strengths of these individual transitions will not be precisely determined in this experiment. However, the individual errors are highly correlated, and in spite of their large value the sum will be known with sufficient precision for our purposes. Shown in Table I are the values of the sum of the 3.96-, 4.05-, 4.07-, and 4.21-MeV transitions resulting from the analysis of the spectra. Also shown is the intensity of the 4.74-MeV transition which may correspond to several primary transitions.

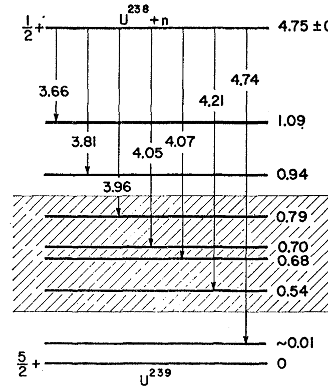

FIG. 5. Level scheme of U^{239} used in the analysis of the resonance-capture spectra. All but the 4.74-MeV transition have been observed in the thermal-capture studies of Fiebiger. The shaded region contains the transitions whose sum was of interest in this and previous experiments. (All energies are in MeV.)

The results in Table I represent the primary experimental data. Two types of statistical analysis of these numbers were made in order to obtain an effective value of ν, the number of degrees of freedom which characterizes the experimental distribution. In each analysis the intensities were treated as the relative values of a partial width corresponding to the sum of the widths of an unknown number of primary transitions. In the first method, proposed by Wilets,[15] the widths for the individual transitions are assumed to be distributed according to the Porter-Thomas distribution ($\nu=1$). An effective value of ν (interpreted as the effective number of open channels) is obtained from the relative variance Δ of the sample of measured widths according to the relationship

$$\nu_{\mathrm{eff}}\equiv\frac{2\langle\Gamma\rangle^2}{\langle\Gamma^2\rangle-\langle\Gamma\rangle^2}=\frac{2}{\Delta^2}\,.$$

Using the results tabulated in Table I gives $\nu=_{\mathrm{eff}}=6.4$

[13] C. C. Rockwood and M. G. Strauss, Rev. Sci. Instr. **32**, 1211 (1961).

[14] G. T. Emery, N. F. Fiebiger, and W. R. Kane, Bull. Am. Phys. Soc. **7**, 722 (1961).

[15] L. Wilets, Phys. Rev. Letters **9**, 430 (1962).

for the 4.0-MeV transition. The value of ν_{eff} was also calculated for the 4.70-MeV transition in this manner, although lack of knowledge of the level structure of U^{239} in this region precludes any detailed interpretation of the result. The value obtained was $\nu_{eff}=6.0$.

For the small population of this experiment (12 resonances), the error in the derived value of ν resulting from the finite size of the sample is much larger than that due to experimental errors in the measurements of the individual partial widths. A second method of determining ν_{eff}, a Monte Carlo procedure developed by Bollinger *et al.*, has the advantage of providing an estimate of this error. For details of the calculation the reader is referred to Ref. 4. Briefly, the procedure consists in generating mathematical samples similar to the experimental sample from a population governed by a χ^2 distribution of a known number of degrees of freedom ν_0. The maximum-likelihood values of ν for these mathematical samples are calculated and then compared with the corresponding ν for the experimental sample. From this comparison for different values of ν_0, a range of values of ν_0 consistent with the data is determined. This procedure was used to estimate ν_{eff} for the relative widths corresponding to the 4.0-MeV band of transitions. The results are shown in Fig. 6, where the probability that the maximum-likelihood value of ν is larger than the value observed for the experimental sample is plotted for various values of ν_0. The result of this calculation is $\nu_{eff}=5.8\pm2.3$.

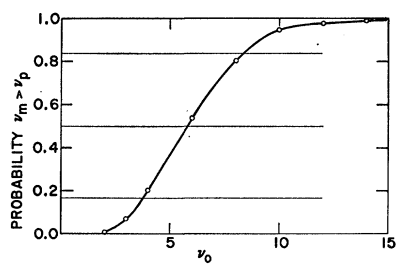

FIG. 6. Summary of results of the Monte Carlo calculation. The number ν_p is the maximum-likelihood value of the number of degrees of freedom for the experimental sample, and ν_m is the corresponding value for the mathematical samples which are generated from a population whose known number of degrees of freedom is ν_0.

IV. DISCUSSION

There has recently been much discussion of possible deviations from the simple statistical model as applied to reaction widths. This has been motivated in part by the early results for the distribution of partial widths in U^{239}. For a group of transitions of equal average strength with no correlation between the individual partial widths, this model would predict a ν_{eff} equal to the number of transitions. But a value of ν_{eff} less than the number of transitions could be interpreted within the framework of the model as evidence for positive correlations between the partial widths. The simple theory does state that negative correlations should not occur, and consequently a ν_{eff} significantly larger than the number of transitions would represent a definite anomaly. Recently, Rosenzweig[16] emphasized that the reported results for U^{238} represent such an anomaly; if they were correct, the simple theory would have to be modified to account for them. Thus, the older results for U^{239} (if accepted) would be quite puzzling, since the statistical model would be expected to apply to U^{239} if it is valid for any nucleus.

Aside from any interpretation, our result for the 4.0-MeV band of transitions, $\nu=5.8\pm2.3$, is in strong disagreement with the results of earlier experiments mentioned in the Introduction. The discrepancy no doubt results from an improvement of experimental techniques, methods of analysis, and more accurate knowledge of the nuclear structure of U^{239}. With regard to interpretation, it is reasonable to assume that all the primary transitions have been observed in the work of Fiebiger, or possibly one has been missed. Thus, in contrast to earlier experiments, our result is consistent with the measured level structure of U^{239}, the results reported for the statistical behavior of partial radiation widths in other nuclei, and the simple statistical theory. More specifically, four primary transitions with uncorrelated partial widths distributed according to a Porter-Thomas distribution adequately explain the observed spectra.

ACKNOWLEDGMENTS

It is a pleasure to acknowledge helpful suggestions of and discussions with Dr. L. M. Bollinger and Dr. N. Rosenzweig.

[16] N. Rosenzweig, Phys. Letters **6**, 123 (1963).