Distribution of Partial Radiation Widths*

L. M. BOLLINGER, R. E. COTÉ, R. T. CARPENTER,† AND J. P. MARION
Argonne National Laboratory, Argonne, Illinois

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The gamma-ray spectra that result from the capture of neutrons in resonances of Hg^{199}, Pt^{195}, W^{183}, and Se^{77} are measured. A least-squares fitting of the spectra gives relative values of partial widths for various sets of high-energy radiative transitions. These widths are treated as statistical samples drawn from populations governed by a χ^2 distribution with ν degrees of freedom. A technique of hypothesis testing that makes use of Monte Carlo calculations is used to derive unbiased values of ν from the small samples of experimental widths. The over-all result of the analysis is $\nu=1.34\pm0.33\pm0.21$. The previously reported partial radiation widths for resonances of Gd^{155}, Yb^{173}, Hf^{177}, and Hg^{201} are also analyzed. The result is $\nu=1.14\pm0.44\pm0.21$. Thus, both sets of data are in good agreement with the value $\nu=1$ that is expected from the Porter-Thomas description of the distribution of the widths associated with a single exit channel.

I. INTRODUCTION

THE measurement and interpretation of the distributions that govern the partial widths of neutron resonances has been one of the most significant contributions slow-neutron spectroscopy has made to nuclear physics. Until recently, the experimental effort has been directed principally at the study[1,2] of the widths for elastic scattering, fission, and radiative capture. Radiative capture is usually a very complex process; its partial width is formed by contributions from many modes of decay. Thus, the distribution of widths for a *single* mode of decay is also of interest. Recent advances in experimental technique have now made it feasible to attempt to measure distributions of this kind. Specifically, we may now attempt to determine the distribution of the partial widths Γ_{rj} for radiative transitions to some single final state j from the series of initial states r formed by capture of neutrons in resonances of a particular spin and parity. For technical reasons the study is still limited to those transitions that proceed directly from the initial state to a final state of low energy. It is the characteristics of these high-energy transitions that are the subject of this paper.

A theoretical background for an understanding of the interest in the distribution of partial radiation widths is provided in the paper by Porter and Thomas,[3] a paper that is devoted primarily to a discussion of the distribution of neutron widths of s-wave resonances. The extremely broad distribution of the neutron widths is shown to result in a natural way from the complexity of the highly excited states formed by neutron capture. The wave functions describing neighboring states of the same spin and parity are assumed to be complex, independent, and random in a way that is intuitively reasonable. Then, from purely statistical considerations, it follows that the matrix elements for neutron emission by way of a single exit channel are distributed approximately normally with a mean value of 0. As a consequence, the reduced neutron widths Γ_n^0 are governed by a frequency function of the form $x^{-1/2}e^{-x/2}$, where $x=\Gamma_n^0/\bar{\Gamma}_n^0$. This distribution, which is often termed the Porter-Thomas distribution, is found to be in good agreement with the experimental values of the neutron widths.[1-3]

As a further consequence of the assumed statistical nature of the wave function of the initial state, it is expected that the widths of a process which can proceed with equal probability by way of ν-independent channels will be governed by a χ^2 distribution with ν degrees of freedom, that is, by a distribution of the form

$$\rho(x,\nu)dx \propto x^{(\nu/2)-1}e^{-\nu x/2}dx. \quad (1)$$

Thus, it has become customary to fit the experimental values of partial widths to the χ^2 distribution and to interpret the derived value of ν as a measure of the number of exit channels available to the process being considered. The results of an impressively large body of experimental data can be summarized with the following statements: $\nu=1$ for the reduced neutron widths, ν is a small number (2 to 4) for fission widths of the common fissile nuclides, and ν is typically a large number for the *total* radiation widths.

In contrast to the generally harmonious experimental evidence concerning the distributions of the total widths for scattering, fission, and radiative capture by slow neutrons, measurements of the partial radiation widths have been in serious disagreement. Since a radiative transition directly to a particular final state appears to be a reaction proceeding by way of a single exit channel, the elementary theoretical considerations of Porter and Thomas lead to the expectation that the distribution of widths for the process would be characterized by $\nu=1$. The first experiment designed to compare partial radiation widths for transitions from initial states formed by neutron capture in different resonances

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† Present address: State University of Iowa, Iowa City, Iowa.

[1] L. M. Bollinger, in *Nuclear Spectroscopy*, edited by F. Ajzenberg-Selove (Academic Press Inc., New York, 1960), Part A, p. 417.

[2] J. A. Harvey in *Neutron Time-of-Flight Methods*, edited by J. Spaepen (EURATOM, Brussels, 1961), p. 23.

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

having the same total angular momentum J was performed by Kennett *et al.*[4] They found the high-energy spectra for two resonances of Mn^{55} to be very different; the relative widths for primary transitions to the second excited state of Mn^{56} differ by at least a factor of 5. This suggests that the partial widths have a broad distribution. In qualitative agreement with this interpretation, Bird[5] reported large differences in the high-energy spectra resulting from capture in three resonances of Hg^{199} with $J=1$. In apparent contradiction to these indications that partial radiation widths are consistent with the expected broad distribution corresponding to $\nu=1$, Hughes *et al.*[6] reported that the widths for transitions to the ground state from the states formed by capture in five resonances of W^{183} deviate from the mean value by only about 20%. This result was interpreted as an indication that the distribution of widths is narrow. On the other hand, in a preliminary report on some of the measurements described in the present paper, Bollinger *et al.*[7] came to the conclusion that ν is a small number and that the experimental data are not inconsistent with $\nu=1$. Moreover, it was shown[7] that at least a part of the apparent uniformity of the widths reported by the Brookhaven group[6] resulted from their failure to resolve single transitions.

The data outlined above and other results of a similar nature[8–11] were brought into sharp focus at the time of the Topical Conference on the Neutron Capture Reaction,[12] Los Alamos, 1959. Some of those present at the Conference felt that the experimental data indicated that the distribution in partial radiation widths was anomalously narrow, a point of view that was ably supported by Hughes[13] in his survey paper. However, the authors of the present paper were led to the conclusion that the apparent uniformity of some of the reported widths probably resulted from experimental factors that discriminate against observation of the small widths that occur frequently in a distribution with $\nu=1$. In particular, there appeared to be a need to avoid the following kinds of systematic errors: (a) Distortion of spectra by an accidental summing of pulses from two gamma rays; (b) a measurement of gamma-ray spectra with too few pulse-height channels;

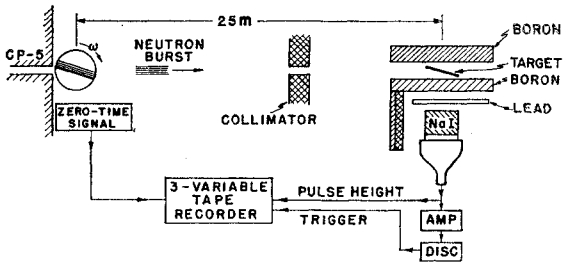

FIG. 1. Experimental apparatus.

(c) deducing radiation widths from the necessarily complex and only partially resolved gamma-ray spectra without a sufficiently quantitative and objective treatment; (d) selecting data on the basis of criteria that tend to discriminate against small widths, and (e) inadequate statistical techniques by which conclusions about the nature of distributions are deduced from the small number of partial radiation widths available. It is significant that *all* of these errors, which are present to some degree in all measurements, tend to inhibit one from obtaining a value of ν as small as 1.

In the present study we have attempted to minimize the pitfalls listed above by striving to obtain gamma-ray spectra of the highest quality, by using quantitative methods of spectral and statistical analysis, and by limiting our measurements to spectra which are simple enough to be interpreted without serious ambiguity. As a consequence of the latter condition, the primary set of measurements have been restricted to high-energy $E1$ transitions from 1^- states formed by neutron capture in the low-energy resonances of even-proton odd-neutron target nuclides with spin $\frac{1}{2}$ and negative parity. The requirement of an even-odd target nucleus results in a compound nucleus for which the low-energy states are rather well understood and the requirement of spin $\frac{1}{2}$ and negative parity for the target permits the spin of the initial compound state to be inferred (though with some uncertainty) from the observed spectra.[14] The nonfissionable nuclides (excluding light nuclides) known to satisfy all of our conditions are Se^{77}, Yb^{171}, W^{183}, Os^{187}, Pt^{195}, Hg^{199}, and Pb^{207}. Of these, we choose not to use Yb^{171} because the spacing of the two lowest states of Yb^{172} is too small and we cannot use Os^{187} and Pb^{207} because at most only one resonance can be isolated with the resolution available. Thus, the targets were limited to Se^{77}, W^{183}, Pt^{195}, and Hg^{199}.

In addition to the four cases just listed, there are many other target nuclides for which the resonant-capture gamma-ray spectra might be expected to yield information about the distribution of partial radiation widths, although with somewhat greater systematic uncertainties. The most favorable of these targets are other even-Z, odd-N nuclides. Fortunately, Carpenter[15]

[4] T. J. Kennett, L. M. Bollinger, and R. T. Carpenter, Phys. Rev. Letters **1**, 76 (1958).
[5] J. R. Bird, in *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958* (United Nations, Geneva, 1959), Vol. 14, p. 294.
[6] D. J. Hughes, M. K. Brussel, J. D. Fox, and R. L. Zimmerman, Phys. Rev. Letters **2**, 505 (1959).
[7] L. M. Bollinger, R. E. Coté, and T. J. Kennett, Phys. Rev. Letters **3**, 376 (1959).
[8] M. K. Brussel and R. L. Zimmerman, Bull. Am. Phys. Soc. **4**, 472 (1959).
[9] J. R. Bird, M. C. Moxon, and F. W. K. Firk, Nucl. Phys. **13**, 525 (1959).
[10] J. R. Bird and J. R. Waters, Nucl. Phys. **14**, 212 (1959).
[11] J. E. Draper and C. O. Bostrom, Nucl. Phys. **14**, 693 (1959).
[12] Abstracts of papers given at the conference appear Bull. Am. Phys. Soc. **4**, 472 (1959).
[13] D. J. Hughes, Brookhaven National Laboratory Report BNL-4464, 1959 (unpublished).
[14] H. H. Landon and E. R. Rae, Phys. Rev. **107**, 1333 (1957).
[15] R. T. Carpenter, Argonne National Laboratory Report ANL-6589, 1962 (unpublished).

has already studied and reported on many of these from a different point of view; hence, for the present paper, we can merely select and use those of his data that are most suitable for our purpose.

Since the time of the Los Alamos Conference, the experimental situation has improved in that there is now general qualitative agreement that ν is a small number for the kind of nuclides considered in the present paper.[16–19] However, as will be seen in later sections, a quantitative comparison of the results from the various laboratories shows that there are still serious discrepancies with regard to the widths of particular transitions and also in the derived values of ν. Moreover, there are conflicting interpretations of the apparent uniformity of the partial radiation widths for the resonances[20–22] of U^{238}, a target for which the spectra are considered to be too complex for inclusion in this paper. Consequently, there appears to be a continuing need for experimental refinement and this need leads us to give a tedious amount of experimental detail in this paper.

II. APPARATUS

An over-all view of the apparatus used in our study of partial radiation widths is given in Fig. 1. Timed neutrons from the Argonne fast chopper[23] are allowed to impinge on the target under investigation. Gamma rays resulting from the capture of these neutrons are detected by a NaI(Tl) scintillation spectrometer. The time of flight of a neutron, as determined from the difference in time between the formation of the neutron burst at the chopper and the detection of a capture gamma ray by the scintillator, is used as a measure of the neutron energy. When certain arbitrary conditions on the pulse height and on the neutron flight time are met, the information is accepted for spectral analysis in a three-parameter tape-recording pulse analyzer.

In all of the measurements, the high-intensity rotor[24] No. III of the fast chopper was used to form the burst of timed neutrons. The rotor was usually operated at a speed of 15 000 rpm to give a burst about 1.5 μsec in width. Thus, with a 25-m flight path, the over-all resolution of the system was about 0.10 μsec/m.

Each target studied consisted of a large slab of the element containing the nuclide of interest. The slab was mounted with the normal to its surface at an angle of about 70° with respect to the direction of the incident beam. The collimation of the beam was adjusted so that the illuminated area of the target was about the same size as the front face of the scintillator. The target was separated from the scintillator by $\frac{1}{4}$ in. of lead and a thick wall of a neutron-absorbing material. These absorbers shield the scintillator from all slow neutrons and from some low-energy gamma rays that originate in the target but have little influence on the high-energy gamma rays.

The primary data of the experiment were pulse-height spectra observed with a single large (15×20 cm) NaI(Tl) scintillator. The resolution width of this spectrometer was about 8.5% for Cs^{137} gamma rays and about 3.5% for 8-MeV gamma rays. The mounting and other characteristics of the large crystal have been described by Managan.[25] In addition to the singles spectra, for some of the targets the spectra associated with coincident pulses in two large scintillators were recorded and used to give auxiliary information of various kinds.

Since some of the measurements required runs as long as 100 h in duration, it was of utmost importance that the pulse height of the detection system should be exceptionally stable. To help achieve this objective, the temperature of the crystal was held constant to within 1°C; and the temperature of all electronic apparatus was held constant to within about 4°C. Also, the photomultipliers were operated at low voltage. Under these conditions, once the system had stabilized at a given counting rate, the pulse height was typically constant to within about $\frac{1}{4}$% over a 24 h period. Thus, the drift in pulse height was negligibly small in many runs. Moreover, if a significant drift did occur, its effect could be eliminated to a large extent by taking the drift into account in the formation of a composite spectrum from independent spectra that were recorded periodically throughout the course of each long run.

The three-parameter tape-recording pulse analyzer that was used to store the spectral information has already been described in detail by Rockwood and Strauss[26] and by Bollinger.[27] It consists of two major units—a data-recording unit and a data-sorting unit. The basic function of the data-recording unit is to store

[16] J. Julien, C. Corge, V. D. Huynh, F. Netter, and J. Simic, J. Phys. Radium **21**, 423 (1960).
[17] L. M. Bollinger, R. E. Coté, and J. P. Marion, Bull. Am. Phys. Soc. **6**, 274 (1961).
[18] F. D. Brooks and J. R. Bird in *Neutron Physics*, edited by M. L. Yeater (Academic Press Inc., New York, 1962), p. 109.
[19] R. E. Chrien, H. H. Bolotin, and H. Palevsky, Phys. Rev. **127**, 1680 (1962).
[20] D. J. Hughes, H. Palevsky, H. H. Bolotin, and R. E. Chrien in *Proceedings of the International Conference on Nuclear Structure, 1960*, edited by D. A. Bromley and E. W. Vogt (University of Toronto Press, Toronto, and North-Holland Publishing Company, Amsterdam, 1960), p. 771.
[21] H. E. Jackson and L. M. Bollinger, Bull. Am. Phys. Soc. **6**, 274 (1961).
[22] C. Corge, V. D. Huynh, J. Julien, J. Morgenstern, and F. Netter, J. Phys. Radium **22**, 722 (1961).
[23] L. M. Bollinger, R. E. Coté, and G. E. Thomas, in *Proceedings of the Second United Nations International Conference on Peaceful Uses of Atomic Energy, Geneva, 1958* (United Nations, Geneva, 1958), Vol. 14, p. 239.
[24] L. M. Bollinger, R. E. Coté, and G. E. Thomas, Argonne National Laboratory Report ANL-6169, 1960 (unpublished), p. 1.
[25] W. W. Managan in Proceedings of the Total Absorption Gamma-Ray Spectrometry Symposium, Gatlinburg, Tennessee, 1960 (TID-7594) (unpublished), p. 120.
[26] C. C. Rockwood and M. G. Strauss, Rev. Sci. Instr. **32**, 1211 (1961).
[27] L. M. Bollinger, in *Proceedings of the Conference on Utilization of Multiparameter Analyzers in Nuclear Physics, Grossinger's, New York, 1962* (U. S. Atomic Energy Commission Report NY 10595), p. 59.

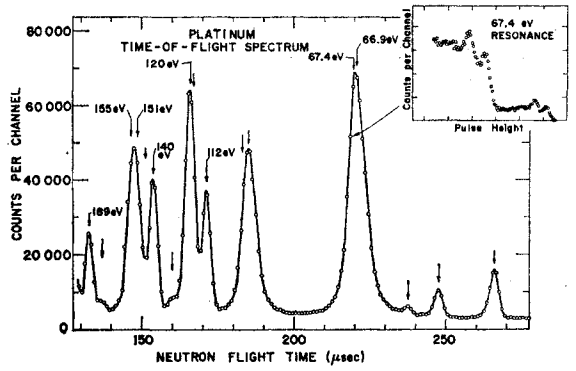

FIG. 2. Representative spectra obtained with a platinum target. The arrows above the main curve show the positions of resonances that are known to be present. Those for which the energy is given are in Pt^{195}.

all useful information from an experiment involving three experimental variables by writing on magnetic tape the measured values of the variables associated with each *individual event* accepted for consideration. At the completion of the measurement, the magnetic tape containing the description of the individual events is transferred to the sorting unit. Here the data are prepared for analysis by sorting the data in such a way as to form the spectra of one variable when arbitrary conditions on the other two variables are satisfied.

An example of the kind of spectra that were recorded for the present study is given in Fig. 2. The main part of the figure shows the intensity of γ-ray counts from a platinum target as a function of neutron time of flight. Associated with each point on this time-of-flight spectrum is a pulse-height spectrum, one of which is given on the figure. Numerous other examples of spectra obtained with our apparatus have been given in Ref. 15.

III. ANALYSIS OF GAMMA-RAY SPECTRA

A. General Method

The primary data of our measurements are pulse-height spectra of a NaI(Tl) scintillator whereas the desired results are the intensities of gamma rays. The capture gamma-ray spectra are so complex for the nuclides studied that the main body of each pulse-height spectrum cannot give useful information about individual gamma rays. However, the first few states of lowest energy in the compound nucleus are sufficiently separated that the high-energy transitions to these states are partially resolved; that is, the shape of the high-energy portion of a pulse-height spectrum depends significantly on the intensities of individual primary transitions to the low-energy states. Consequently, the shape of a pulse-height spectrum should enable one to deduce the intensities of transitions to the first few states.

The basic physical assumption in the analysis of the pulse-height spectra is the customary one that each "complex" spectrum is formed by a linear superposition of a set of simple spectra which, in our case, are individual γ-ray lines. Then, for the most elementary complex spectra, in which the position and shape of each contributing line is known, the intensity a_{rj} of a transition from an initial state r to a final state j can be determined by making a linear least-squares fit of the experimentally measured pulse-height spectrum to a relationship of the form

$$y_{ir} \equiv (N_{ir} - B_{ir}) = \sum_j a_{rj} R_{irj}, \tag{2}$$

where N_{ir} is the total number of pulses falling within the ith channel, B_{ir} is the number of background counts associated with the ith channel, and R_{irj} is the contribution to the ith channel from a transition $r \rightarrow j$ of unit intensity.

The procedure used to obtain the least-squares fit is well known.[28] First, we find the least-squares values of the intensities a_{rj} as those values that minimize the variance V_r defined by the relationship

$$V_r = \sum_i w_{ir} (y_{ir} - \sum_j a_{rj} R_{irj})^2, \tag{3}$$

where w_{ir} is the weight attached to the measurement at each channel i. The weights are ordinarily set equal to $(\Delta y_{ir})^{-2}$, the quantity Δy_{ir} being the standard statistical error of y_{ir}. Then, one obtains the statistical uncertainty in the intensities from the relationship

$$(\Delta a_{rj})^2 = \frac{\chi_r^2}{n-1} (H_r)_{jj}^{-1}, \tag{4}$$

where the $(H_r)_{jj}^{-1}$ are the diagonal elements of the inverse matrix defined by

$$(H_r)_{jm} = \sum_i w_{ir} R_{irj} R_{irm}.$$

Here n is the number of degrees of freedom in the fit and χ_r^2 is the minimum value of V_r. However, χ_r^2 is itself a random variable subject to statistical fluctuations. Thus when χ_r^2 is less than its expectation value, the latter value is used in Eq. (4). As a final step, the quality of the fit is tested by comparing the experimental value of χ^2 with the probability distribution that is expected for a least-squares fit with a known number of degrees of freedom. Failure of the experimental value to be consistent with the theoretical distribution is a strong indication that the assumptions entering into the fit are not all valid or that systematic errors are present in the measurement.

Although the simple least-squares procedure outlined above is the basic one used for spectral analysis, it requires considerable amplification to make it a con-

[28] M. G. Kendall and A. Stuart, *Advanced Theory of Statistics* (Charles Griffin and Company Ltd., London, 1961), Vol. 2, Chap. 19.

venient and sensitive method for analyzing capture gamma-ray spectra. Perhaps the most serious difficulty concerns the position of the individual gamma-ray lines in the complex spectrum. As was stated above, the position of these lines must be known if we are to make use of the linear least-squares method. However, in the capture gamma-ray spectra we often do not know the energy or even the number of the lines contributing to a particular range of the spectrum. Moreover, even when the *spacing* between all lines is known, the *position* of the lines relative to the positions of the reference lines is rarely known with sufficient accuracy. To overcome this difficulty, the line positions are found in basically the same way as are the intensities, namely, by finding the set of line positions that results in the minimum value of χ^2. However, since it is not feasible to perform the minimization analytically, we make use of the iterative technique of variable metric minimization.[29] Known physical information about the spectra, such as the spacing between lines, is used to place constraints on the line positions. If the number of lines as well as their positions are unknown, the best that can be done is to determine the minimum number of lines that gives an acceptable value of χ^2.

A special characteristic of the resonant-capture gamma-ray spectra is that, because the neutron resonances are so close together, effectively the same set of gamma-ray energies is present in all the spectra of a given nuclide. Moreover, not only are the energies the same but, since the spectra are all recorded simultaneously in the three-parameter analyzer, the positions of the lines in the experimental data are also the same. It is clear that this physical information should be used as a constraint in the analysis. This objective is achieved by performing the minimization for all spectra simultaneously. That is, all the intensities a_{rj} and energies E_j for the set of spectra are obtained simultaneously by minimizing the over-all variance

$$V(a_{11},\cdots,E_1,\cdots)=\sum_r \sum_i w_{ir}[y_{ir}-\sum_j a_{rj}R_i(E_j)]^2, \quad (5)$$

where E_j is the energy of a transition to the state j from any of the initial states r and the subscript r on R is dropped to indicate that the line shapes are the same for all resonances. The errors associated with the a_{rj} should include a term to take account of the uncertainties in the line positions. However, since this effect is usually unimportant (but not always), for mathematical convenience we continue to calculate errors with an expression of the same form as Eq. (4).

The variance [Eq. (5)] is minimized on an IBM-704 computer. The reference line shape R_{ij} for any given energy E_j is formed by linearly interpolating between tabulated shapes for two or more gamma-ray energies. Thus, all the derivatives that are required for the determination of the parameters and their statistical errors must be obtained by numerical methods. The largest problem that can be treated is one in which there are ten spectra formed by ten gamma-ray lines, with each spectrum specified by 50 points.

[29] W. C. Davidon, Argonne National Laboratory Report ANL-5990 Rev., 1959 (unpublished).

B. Experimental Problems

This paper is mainly concerned with the intensities of transitions to a few low-energy states that are thought to be well isolated from other states. For these intensities, the principal sources of systematic error that need to be considered result from uncertainties in the true line shapes and in the background corrections. These uncertainties could be important for this experiment because they make it difficult to determine the intensities of weak transitions, and these weak transitions would be very frequent in a population governed by a χ^2 distribution with one degree of freedom. In this section we discuss the methods that were used to determine the line shape and the background.

Because of the limited range of gamma-ray energy that is being treated, the energy dependence of the line shape is relatively unimportant. The important problem is to determine an accurate shape for a single gamma-ray energy falling in the range from 7 to 8 MeV. A possibly useful source for this purpose is the gamma radiation resulting from capture of thermal neutrons in aluminum. The high-energy portion of the spectrum is dominated by a single line at 7.7 MeV. However, because of the small neutron-capture cross section of aluminum, the capture gamma-ray spectrum was found to be accompanied by a background spectrum that is intense enough to introduce a significant uncertainty in the true line shape. A more convenient and reliable measure of the line shape is obtained from the neutron-capture gamma-ray spectrum of iron. This spectrum is complex,[30] but a line[31] at 7.64 MeV is much stronger than any other line in the energy range of interest to us. Thus, since all the lines with a significant intensity have been resolved and since their relative intensities are known, the shape can be obtained by substituting the known y_i and a_k into Eq. (2).

An example of the capture gamma-ray spectrum of iron is given in Fig. 3. The spectrum was observed with a beam of neutrons filtered through 20 cm of bismuth. A beam of this kind has a mean neutron energy of about 0.0015 eV and is almost entirely free of gamma rays and fast neutrons. The reference line shape obtained from the iron spectrum is shown by the dashed line of Fig. 3. It is seen to have the desirable property that the full-

[30] L. V. Groshev, V. N. Lutsenko, A. M. Demidov, and V. I. Pelekhov, *Atlas of Gamma-Ray Spectra from Radiative Capture of Thermal Neutrons*, translated by J. B. Sykes (Pergamon Press Ltd., London, 1959).

[31] This line actually is formed by two components, as demonstrated by N. F. Fiebiger, W. R. Kane, and R. E. Segel, Phys. Rev. 125, 2031 (1962). However, the difference in the energies of the two gamma rays is so small (14 keV) that no significant error results from treating the pair of lines as a single line.

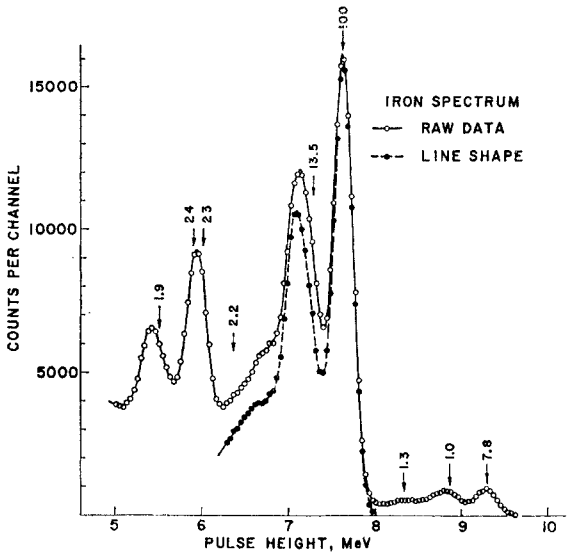

FIG. 3. Pulse-height spectrum from capture of thermal neutrons in iron. The arrows indicate the energies and the associated numbers indicate the relative intensities of lines that are known to be present in the spectra. The solid points define the line shape that is derived from the spectrum.

energy peak is much more prominent than is the peak associated with the escape of one annihilation quantum and the peak associated with the escape of two quanta is almost undetectably small—properties that result from the large size of the scintillator. Because of the freedom from background and the good statistical accuracy of the data, the principal source of error in the line spectrum is believed to be an uncertainty associated with the relative intensity of the line at 7.28 MeV. Groshev *et al.*[30] give a value of 16.8 (relative to a value 100 for the 7.64-MeV line) whereas Bartholomew and Higgs[32] give a value of 10.3 for the intensity. We use the mean of these two values. The line shape obtained with an iron target in this way is in excellent agreement with the one obtained with an aluminum target. The accuracy of the shape is discussed further in Sec. V.

Although the line shape derived from the 7.64-MeV gamma ray in the iron spectrum was the most important one used, for some analyses the energy dependence of the line or the shape in an entirely different range of energy was required. In these cases, the 9.32-MeV gamma ray in the iron spectrum or the spectrum from capture in the 96-eV resonance of Pt^{198} was used. The latter spectrum is formed, as far as one can determine, by just two high-energy lines—a strong line at about 5.48 MeV and a weaker line at about 4.46 MeV.

A careful study was made to determine the behavior of the spectrum from background radiations, a subject that has already been discussed in detail elsewhere.[15] In the range of neutron energy that is of interest here (5 to 1000 eV), it was shown that timed neutrons scattered out of the sample do not give a significant contribution to the background, presumably because these neutrons are absorbed by the thick slabs of B_4C that are placed around the target (Fig. 1). Moreover, the intensity and the shape of the background spectrum from other sources was found to change only slowly with neutron energy. Thus, the background spectrum at any given resonance may be determined by an interpolation between the spectra observed in off-resonance regions in which the capture cross section of the target is known to be small. For the targets considered in this paper, the systematic errors introduced by this procedure of determining the background are believed to be negligibly small.

In addition to subtracting a background of the kind just considered, we must take account of a distortion of spectra by the addition of pulses from more than one gamma ray. This effect may result either from the chance addition (pile up) of pulses from independent sources or from the summing of pulses from the cascade of gamma rays produced by the capture of a single neutron. In all of the measurements reported here, the contribution from pile-up pulses may be neglected. However, for some of the targets the summing of related pulses is an important effect. One does not ordinarily have enough information to calculate the magnitude of the effect; it must be measured. The most elegant way to make the measurement is to record the spectrum of the *sum* of coincident pulses in two scintillators mounted on opposite sides of the target. If these scintillators are of the same size, have the same geometry with respect to the target, and have the same line shapes, the sum-coincidence spectrum has the same shape and contains exactly twice the intensity of sum pulses as does the spectrum of sum pulses in a single oscillator. Also, if the resolution width of the coincidence circuit is adjusted to an appropriate value, as determined from the characteristics of the analog-to-digital converter in the pulse-height analyzer, the intensity of pile-up pulses in the sum-coincidence spectrum is also approximately twice that in the singles spectrum. Thus, by making a sum-coincidence measurement at each resonance, the correction for both summing and pile up can be made with great accuracy. Examples of sum-coincidence spectra have been given elsewhere.[33]

Unfortunately, the two large scintillators required for a sum-coincidence measurement were not available for the study of all of our targets. Thus, it was necessary to devise another and somewhat less satisfactory way to determine the magnitude of the background from sum pulses. The method used depends on the fact that the rate of sum pulses is proportional to the *square* of the effective solid angle of the scintillator with respect to the target, whereas the rate of single pulses varies

[32] G. A. Batholomew and L. A. Higgs, Chalk River Laboratories Report CRGP-784, 1958 (unpublished).

[33] L. M. Bollinger and R. E. Coté, Argonne National Laboratory Report ANL-6146, 1960 (unpublished).

only as the first power. Thus, one can correct for the sum pulses by using the information contained in the spectra obtained in two runs with different solid angles. In practice, the solid angles used in the two runs differed by a factor of about 3, with the front face of the crystal about 23 cm from the center of the sample in one run and about 11 cm in the other run.

IV. CAPTURE GAMMA-RAY SPECTRA

The initial results obtained from the least-squares fits of the experimental spectra are intensities a_{rj}. These incorporate a scale factor proportional to the number of neutrons captured within the time-of-flight interval contributing to each spectrum r. Thus, to reduce all of the a_{rj} for a given target to the same units, it is necessary to divide each a_{rj} by a quantity that is proportional to the number of neutrons captured to form its spectrum. The resulting normalized intensities are labeled A_{rj}. Fortunately, for our purposes it is not necessary to determine the normalizing factors in an absolute way. Consequently, we have adopted the customary expedient of dividing the a_{rj} by some gamma-ray intensity that is believed to be approximately proportional to the number of neutrons captured; typically, the intensity of gamma-ray pulses with energy greater than about 3 MeV was used. The validity of this procedure depends on the assumptions that the multiplicity and the gross features of the capture gamma-ray spectra of a given nuclide are approximately the same for all resonances of a particular spin and parity. The complexity of the capture gamma-ray cascade makes these assumptions reasonable for the nuclides studied. Moreover, the assumptions are supported by the experimental observations that (a) the pulse-height spectra are closely similar from resonance to resonance, except in the high-energy portion which, for the nuclides studied, contributes only a relatively minor part of the total radiation width; (b) in the case of the Hg^{199} resonances, the experimental multiplicities are the same for three resonances within the experimental accuracy of about 10%; (c) the total radiation widths of[34] W^{183} and of[35] Hg^{199} are approximately the same from resonance to resonance; and (d) for Pt^{196}, the ratio of the intensity of all gamma rays to the sum of intensities of the strong lines resulting from decay of the 2^+ states at 354 and 686 keV is approximately the same for the several resonances at which the ratio has been measured.[36]

The principal reason for any doubt about the validity of the assumption that the number of neutrons captured is proportional to the intensity of a broad band of low-energy gamma rays arises from the recent evidence that, for a few special cases, the total radiation width Γ_γ and the gross features of the capture gamma-ray spectrum *do* differ significantly from resonance to resonance. This effect was demonstrated[35] most clearly for Hg^{201}, for which the changes in Γ_γ were shown to be correlated with changes in the intensities of a few exceptionally strong high-energy transitions. The total radiation widths of Pt^{195} (one of the targets studied in this experiment) have also been reported[37] to differ greatly from resonance to resonance. In this case, however, there is no qualitatively obvious correlation between the reported values of Γ_γ and the characteristics of the gamma-ray spectra observed in the present experiment. Moreover, for Pt^{195} the high-energy transitions are much less important, relative to the low-energy transitions, than they are for Hg^{201}. Thus, since the reported fluctuations of Γ_γ for Pt^{195} cannot easily be explained, we feel that there is reason to doubt the validity of the measured values of Γ_γ. In any case, it is not clear that fluctuation in Γ_γ would necessarily lead to significant errors in the normalizing factors obtained from the intensity of gamma rays within a broad band of energy.

In view of the uncertainty about the normalizing factors for the Pt^{195} resonances, we are fortunate to have data of Moxon and Rae[38] that permit the intensity of gamma rays giving pulses in NaI greater than 2 MeV to be compared with the rate of neutron capture. The ratios of these two quantities are found to be 1014 ± 14, 1017 ± 17, 1001 ± 16, 957 ± 23, 999 ± 13, 974 ± 19, 1005 ± 13, and 1026 ± 32 (arbitrary normalization) for the resonances at 12, 19, (66.9+67.4), 112, 120, 140, (151+155), and 189 eV, respectively. Thus, for Pt^{195} the intensity of gamma rays in a broad range of energy is seen to be a reliable measure of the number of neutrons that are captured. In view of the evidence cited in an earlier paragraph, one expects that the gamma-ray intensity is equally meaningful for the resonances of W^{183} and Hg^{199}. It is only for Se^{77}, then, that some doubt remains. A qualitative examination of the spectra of Se^{77} leads one to expect that their behavior would be similar to that of the resonances of Hg^{201}. Thus, on the basis of the results of the previously reported study[35] of Hg^{201} resonances, it is estimated that the normalizing constant used for Se^{77} might be uncertain by about $\pm20\%$. An uncertainty of this magnitude is hardly significant, in view of the order-of-magnitude fluctuations that the partial radiation widths themselves exhibit.

The general procedure used to determine the relative intensities a_{rj} was outlined in Sec. III. Usually the analyses were carried out in two steps. First, a limited range of pulse height was used to obtain the intensities of transitions to the two or three states of lowest energy. The values of χ^2 obtained in these analyses are entirely acceptable in most cases. The second step in each

[34] J. R. Waters, J. E. Evans, B. B. Kinsey, and G. H. Williams, Nucl. Phys. **12**, 563 (1959).
[35] R. T. Carpenter and L. M. Bollinger, Nucl. Phys. **21**, 66 (1960).
[36] H. E. Jackson and L. M. Bollinger, Phys. Rev. **124**, 1142 (1961).
[37] J. Julien, C. Corge, V. D. Huynh, J. Morgenstern, and F. Netter, Phys. Letters **3**, 67 (1962).
[38] M. C. Moxon and E. R. Rae (private communication).

analysis was to fit the experimental spectra over a larger range of pulse height, typically a range thought to include about six transitions. Often the value of χ^2 obtained in fitting the larger range is just barely acceptable. This indication of systematic errors probably results from the presence of unknown transitions, from a failure to subtract the background properly, or from the influence of the tails of transitions that are outside the range of pulse height being considered. This latter effect is a difficult one to take into account objectively. The procedure used was to make the curve fitting include the tails of known or hypothetical transitions of slightly lower energy than the range of energy being treated. Then, if the intensities of the transitions inside the range of consideration proved to be insensitive to the number and positions of the hypothetical transitions outside the range, the intensities were judged to be reliable in the sense that the intensities of strong transitions are approximately correct. All of the intensities listed in Tables I–IV are reliable in this sense. However, it should be recognized that the intensities of weak transitions to the states of higher energy are likely to be in error by amounts that are much greater than the quoted statistical errors. Because of these systematic uncertainties, the intensities of transitions to the higher states are not used in the determination of the distribution of partial radiation widths.

The errors listed in the tables of intensities are all standard statistical uncertainties. The probable magnitude of various kinds of systematic errors are indicated in the text.

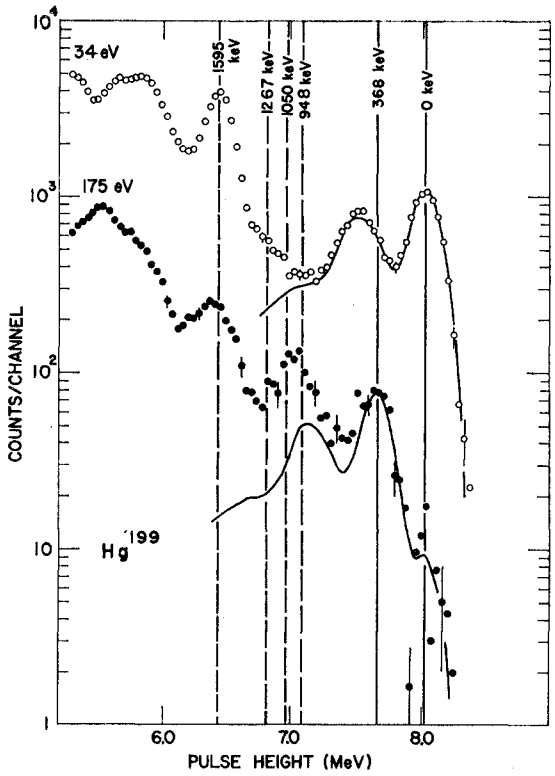

FIG. 4. Pulse-height spectra for resonances of Hg^{199}. The pulse heights corresponding to transitions to known low-energy states are indicated by vertical guide lines. The solid curves through the data points are the least-squares fits made to obtain A_0 and A_1.

A. Hg^{199} Spectra

The target used for the study of the Hg^{199} resonances was HgS in a thickness of about 1.4 g/cm^2. All known resonances[35] of Hg^{199} in the range from 30 to 300 eV were studied, namely, those at 34, 175, and 260 eV with $J=1$ and the one at 130 eV thought to have[39] $J=0$. The magnitude of the contribution from sum pulses was determined by making two runs with different solid angles. Two of the γ-ray spectra observed with the smaller solid angle are given in Fig. 4. The time-of-flight spectrum has been given elsewhere.[15]

The known low-energy states[40,41] of the compound nuclides being studied are shown in Fig. 5. For Hg^{200}, the two states of lowest energy (0 and 368 keV) are seen to be widely enough separated from the next lowest state (948 keV) that the high-energy portions of the capture gamma-ray spectra may be analyzed under the assumption that transitions to the ground state and first excited state are the only contributors. In the following discussion it is convenient to refer to the intensities of transitions to these two final states as a_0 and a_1 (or A_0 and A_1), the subscript r for the initial state being dropped.

In view of the simplicity of the line structure in the spectra for Hg^{199}, the most obvious difficulty associated with their analysis is the correction for sum pulses. Although the ideas for using the spectra recorded with two solid angles (as outlined in Sec. III) are entirely valid in principle, in practice a straightforward calculation to obtain a corrected spectrum gives a result with a much poorer statistical accuracy than the original spectra. Thus, one would prefer not to make the correction but merely to use the results of the two sets of data to show that the correction is unnecessary. This hope turns out to be fulfilled for the Hg^{199} spectra. If sum pulses do make a significant contribution, the shapes of the high-energy sections of the spectra would be expected to depend on the solid angle since the shapes of the sum spectra usually differ greatly from the shapes of the singles spectra. Yet, for all three resonances with $J=1$, a quantitative examination reveals no statistically significant difference in the shapes of the spectra obtained with the two different solid angles. For example, for the 34-eV resonance we obtain the ratio $a_1/a_0=0.060\pm0.018$ from the spectrum measured with the smaller solid angle and obtain 0.069 ± 0.040

[39] For a further discussion of this subject, see the Appendix.
[40] *Nuclear Data Sheets*, compiled by K. Way *et al.* (Printing and Publishing Office, National Academy of Sciences–National Research Council, Washington 25, D. C., 1962).
[41] R. T. Carpenter, R. K. Smither, and R. E. Segel, Bull. Am. Phys. Soc. 7, 11 (1962).

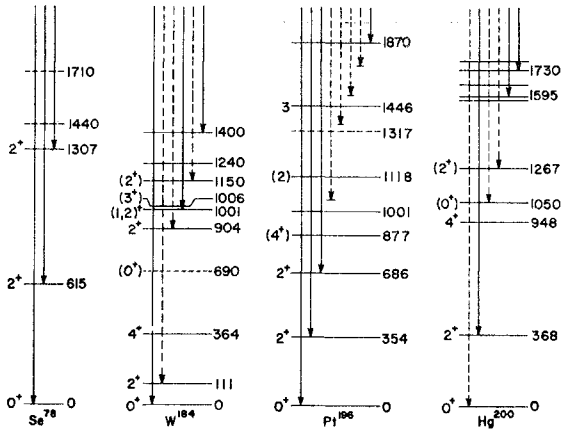

FIG. 5. Energy levels (Refs. 40, 41) of compound nuclides formed by neutron capture. The transitions shown as solid lines have been observed in thermal-neutron-capture gamma-ray spectra (Refs. 30, 32). In addition to these transitions, in resonant capture we observe the transitions shown as dashed lines. All energies are in keV.

after correcting for summing. Thus, the error from sum pulses is entirely obscured by purely statistical uncertainties so that we feel justified in using the intensities obtained from the measurements with the smaller solid angle without correction for sum pulses. These results are given in Table I.

TABLE I. Intensities of high-energy transitions to low-energy states in Hg^{200}. The errors listed are standard statistical uncertainties.

Resonance energy (eV)	Energy of final state (keV) 0	368	948	1050	1267
34	1779±28	106±31	61±43	−45±32	543±38
175	79±22	683±53	188±119	755±103	65±110
264	1142±64	888±102	465±224	110±191	1480±228
Mean	1000	559	238	273	696

In addition to summing, another kind of systematic error needs to be kept in mind, namely, the one that could result from an error in the reference line shapes. For almost all transitions reported in this paper, however, the errors in intensity that result from errors in the line shape are small compared with the statistical uncertainties. This possible source of error will be discussed in detail in Sec. V, which follows presentation of the data.

For the 264-eV resonance, a systematic error could also result from our failure to resolve it from the small unidentified resonance at 271 eV. The error is believed to be negligible, however, since quantitative calculations show that the rate of neutron capture in the 271-eV resonance is only a small fraction of that in the 264-eV resonance.

Up to this point we have restricted our attention entirely to a determination of the intensities of transitions to the ground state and first excited state of Hg^{200}. We should now make sure that there are no previously undetected states of Hg^{200} at a low enough energy to cause errors in the intensities of the transitions to the first two states. Let us therefore extend the analysis to a wider range of energies and inquire whether the spectra can be accounted for in terms of transitions to known low-energy states. For this purpose, the spectra were analyzed by permitting transitions to all of the known states of Hg^{200} that have energies less than 1.3 MeV, namely, states at 0, 368, 948, 1050, and 1267 keV. In each case, a satisfactory fit was obtained in terms of these transitions. Thus, there is no evidence for the existence of new states in Hg^{200} at energies less than 900 keV. This gives us renewed confidence that the intensities of transitions to the first two states are reliable. The intensities that are obtained for transitions to the states of higher energy are given in Table I. Note that here and elsewhere the values of A_0 and A_1 are those obtained from the analysis over a small range of γ-ray energies because these values are believed to be less sensitive to the influence of the various systematic errors than are the results obtained for a wider range of energy. Actually, there is no significant difference between the two sets of values.

B. Pt^{195} Spectra

The target used for the study of the resonances of Pt^{195} was platinum metal with a thickness of about 2.3 g/cm^2. The spectra studied are those for the eight resonances[37] that are at energies less than 200 eV and are thought to have[39] $J=1$. The spectra for resonances at higher energy were not considered because, in view of the close spacing of resonances in normal platinum, it seemed improbable that individual resonances could be resolved with certainty.[39] A time-of-flight spectrum for platinum has been presented in Fig. 2. All of the γ-ray spectra that were accepted for analysis are given in Fig. 6, including the spectrum observed at the 155-eV resonance, for which $J=0$.

The high-energy end of each γ-ray spectrum was analyzed under the assumption that it is formed by direct transitions to the three states of lowest energy in Pt^{196}, namely, those at 0, 354, and 686 keV. The relative widths obtained from these analyses are given in Table II, and the calculated spectra corresponding to these widths are shown in Fig. 6. The fit is seen to be excellent in the range dominated by the three transitions of interest.

The methods used to check for possible systematic errors were the same as those described for the Hg^{199} data. Again comparing the spectra recorded with different solid angles revealed that the errors from pile up and from sum pulses were undetectably small. The principal uncertainties that need to be considered, then, are those that result from imperfect resolution in the neutron time of flight, an effect which influences our interpretation of the data for the resonances at 67.4,

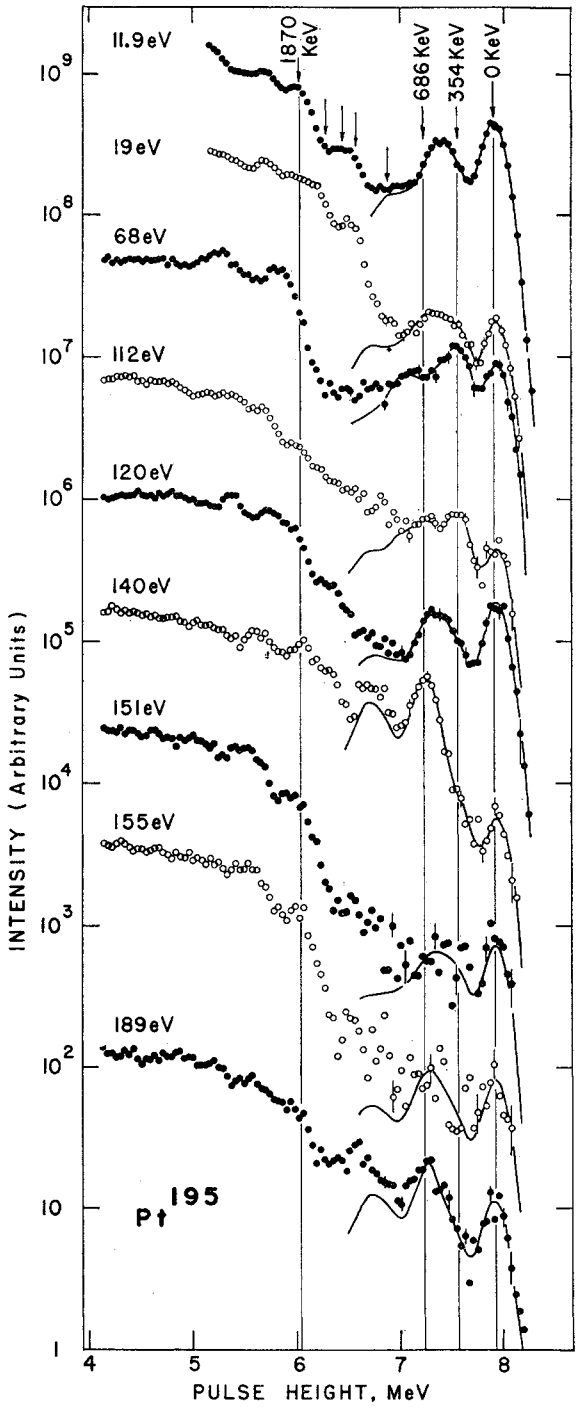

FIG. 6. Pulse-height spectra for resonances of Pt^{195}. The pulse heights corresponding to transitions to known low-energy states are indicated by vertical guide lines. Other transitions suggested by the spectra are shown by arrows alone. The solid curves through the data points show the least-squares fit for the three transitions of highest energy.

120, 151, and 155 eV. The 67.4-eV resonance is known to be accompanied by one at 66.9 eV, a difference in energy that corresponds to a difference of only 0.8 μsec in the time of flight in our experiment. Since the resolution width of the system is about 2.5 μsec, there might be some question about the possibility of making a reliable determination of the spectrum for the 67.4-eV resonance alone. We find that it is possible, however, by using only those data contained in the time-of-flight channel about half-way up the high-energy side of the peak that is formed by the two resonances. The relative intensity of gamma rays in the range from 7 to 8 MeV is observed to be about four times as great for this channel on the high-energy side as is the same intensity for the corresponding time channel on the low-energy side of the peak. Simple but tedious numerical calculations based on this observation show that the 66.9-eV resonance contributes not more than one fifth of the counts to the low-energy portion of the spectrum associated with the point on the high-energy side of the time-of-flight peak. Moreover, since the 66.9-eV resonance has been assigned[36] to the isotope Pt^{195}, both the close spacing of the 66.9- and 67.4-eV resonances and the absence of strong high-energy transitions make it almost certain that $J=0$ for the 66.9-eV resonance. In this case, the 66.9-eV resonance would contribute one-fifth of the low-energy counts and none of the high-energy counts to the spectrum for the channel on the high-energy side of the time-of-flight peak. The values given in Table II were obtained under this assumption. In any case, even if $J\neq 0$ for the 66.9-eV resonance, the systematic error in the partial radiation width would be less than 20%.

TABLE II. Intensities of high-energy transitions to low-energy states in Pt^{196}. The errors listed are standard statistical uncertainties.

Resonance energy (eV)	Energy of final state (keV) 0	354	686	$\sum_{j=0}^{2} A_j(E_0/E_j)^3$
12	2355±24	214±26	39±26	2651
19	433±8	210±10	226±12	970
67.4	2019±52	1782±73	97±71	4184
112	556±25	676±40	353±44	1792
120	1413±33	122±36	452±42	2146
140	275±17	225±28	2495±54	3808
151	246±28	71±37	75±41	425
189	703±42	115±55	943±77	2073
Mean	1000	427	585	

The problem in treating the close pair of resonances at 151 and 155 eV is similar to that just discussed. However, in this case the two resonances are separated in time by 1.9 rather than only 0.8 μsec. Thus, in view of the outcome of the above discussion, the spectrum associated with the time channel on the low-energy side of the time-of-flight peak around 153 eV unquestionably results almost entirely from the single resonance at 151 eV.

In the case of the 120-eV resonance, an uncertainty is introduced by the presence of a narrow resonance about $\frac{1}{2}$ eV lower in energy. Little is known about this

level except that it gives a weak effect, in comparison with the 120-eV resonance, both in transmission experiments and when capture gamma rays are detected. Thus, the influence of the small resonance has been ignored in our analysis.

A somewhat more subtle kind of systematic error needs to be mentioned in connection with the resonances at 19 and 112 eV. As has recently been demonstrated experimentally,[42] under some circumstances the intensities of *individual* transitions are strongly influenced by an interference between the amplitudes contributed by two or more resonances. Thus, it may be of importance to observe spectra that are associated with time channels that are accurately centered around the resonance energy. A failure to do so could easily lead to errors of about a factor of 2 in the intensities of the transitions for the 19 and 112-eV resonances, since the relatively weak high-energy transitions for these resonances interfere strongly with the much more intense transitions for the nearby resonances at 12 and 120 eV, respectively.

As a final check on the reliability of the analyses described above, let us search for evidence for the presence of low-energy states in Pt^{196} that have not been reported previously. A qualitative examination of Fig. 6 shows immediately that there are several such states. A quantitative treatment shows that at least four low-energy states are required in the energy range from 700 to 1500 keV and, if only four states are present, they are in the neighborhoods of 1060, 1350, 1500, and 1650 keV. However, the quality of the fit obtained with transitions to these four states is too poor to give one any confidence that the fit is a unique description of the spectra. In any case, there is no evidence for new states at a low enough energy to influence the values of the intensities of the three transitions of highest energy.

C. W^{183} Spectra

The target used for the study of the W^{183} resonances was tungsten metal about 1.9 g/cm² thick. The measurements covered the resonances at 7.6, 27, 41, 46, and 65 eV, all of which are known[34] to have $J=1$. No effort was made to study the resonances at higher energy because of the difficulty of resolving single resonances with certainty. Preliminary measurements had shown that the intensity of sum pulses from W^{183}, unlike those from Hg^{199} and Pt^{195}, was great enough to cause small but statistically significant errors. Thus, the contribution from sum pulses was determined by a sum-coincidence measurement.

The known states of W^{184} are shown in Fig. 5. Transitions to the 4^+ state at 364 keV would be expected to be negligibly weak and in fact could not be detected in our spectra. Hence, the high-energy end of each spectrum was treated by considering transitions only to the ground state and the first excited state at 111 keV. Because the difference in energy between these states is only about half a resolution width, the two transitions to the states form what appears to be a single high-energy line. Consequently, a rather refined analysis is required to determine the individual intensities a_0 and a_1. On the other hand, the sum (a_0+a_1) can be determined with ease, since the sum is closely related to the intensity of the apparently single line. The results obtained for the sum (A_0+A_1) are listed in Table III.

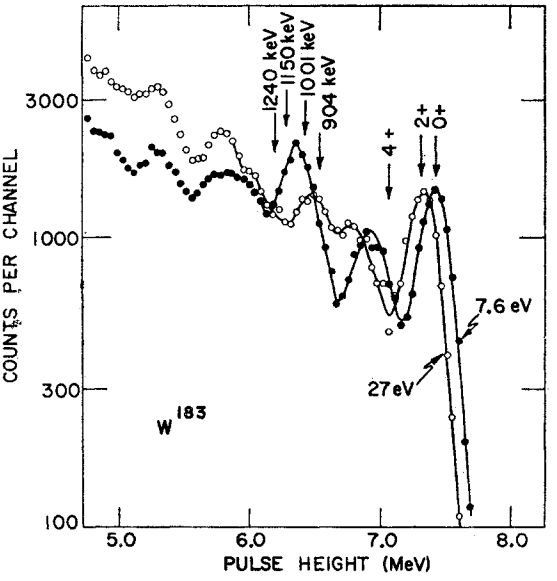

FIG. 7. Pulse-height spectra for resonances of W^{183}.

The determination of the intensities of the individual transitions makes use of the fact that the *position* of the apparently single line in the complex spectrum depends on the ratio a_1/a_0. Implementation of this idea requires that the position of the reference line should be accurately known relative to those of the individual lines in the complex spectrum. The required accuracy usually cannot be achieved when the line shape is obtained from a source that is independent of the target being studied. Fortunately, however, the spectra of two of the W^{183} resonances, those at 7 and 27 eV, happen to have characteristics that permit an accurate determination of the position as well as the shape of the reference line. Comparing the high-energy ends of these spectra, which are given in Fig. 7, we observe that the 7-eV spectrum is displaced significantly to higher energies relative to the 27-eV spectrum. To a first approximation, this displacement is a rough measure of the quantity $[(a_1/a_0)_7+(a_0/a_1)_{27}]$; the observed displacement implies that this quantity is about 0.08. Thus, we see immediately that $(a_1/a_0)_7$ is small and that $(a_1/a_0)_{27}$ is large.

We now seek independent values of the two ratios a_1/a_0. First, let us fit the spectra with a line *shape* that is obtained from the iron spectrum but a line *position*

[42] R. E. Coté and L. M. Bollinger, Phys. Rev. Letters 6, 695 (1961).

TABLE III. Intensities of high-energy transitions to low-energy states in W^{184}. The errors listed are standard statistical uncertainties.

Resonance energy (eV)	Energy of final state (keV) 0	111	690	904	1001	1150	(A_0+A_1)
7	2775±132	86±132	119±46	129±86	2019±119	1511±81	2855±24
27	63±95	1580±95	−56±26	691±44	321±56	338±34	1643±16
41	89±90	730±90	49±56	162±95	561±129	387±84	819±30
46	1063±90	420±90	46±25	6±40	965±56	89±33	1483±18
65	1005±140	1502±150	−29±127	−324±196	821±288	2196±196	2507±73
Mean	1000	862	26	133	937	904	

that is treated as a free parameter. Satisfactory fits are obtained with pairs of ratios in the ranges $(a_1/a_0)_7 = (0 \text{ to } 0.10)\pm0.03$ and $(a_0/a_1)_{27} = (0.08 \text{ to } 0)\pm0.04$. The errors listed are the statistical errors corresponding to a definite line position and the range of values reflects the range of line positions that yield a satisfactory value of χ^2. In this case, the dominant source of error is seen to be the uncertainty in the line position.

In an effort to minimize the influence of the uncertainty in line position, let us now use an alternative procedure in which a reference line shape is deduced from the complex spectra themselves. We proceed by deriving line shapes from the 7-eV spectrum with various trial values of $(a_1/a_0)_7$; in this way, for a given value of $(a_1/a_0)_7$ the line position is completely determined. Then when these line shapes are used in the analyses, other spectra can be satisfactorily fitted with ratios in the range $(a_1/a_0)_7 = (0 \text{ to } 0.15)$ and $(a_0/a_1)_{27} = (0.04 \text{ to } 0)\pm0.05$. This result is in good agreement with that obtained with the iron line but the errors are still large.

The results for the intensities A_0 and A_1 are summarized in Table III, where the errors given include the effect of the uncertainty in the line position. Note that these values of intensity are in good agreement with those obtained by us previously[7] with a much less powerful experimental system.

The final step in the analysis of the W^{183} spectra is, as usual, to search for evidence of states that might tend to invalidate the values of a_1/a_0 obtained by considering only two transitions. For this purpose, the part of each spectrum at energies greater than about 6.1 MeV were analyzed in terms of transitions to all the known states of W^{184} below 1290 keV. This range of energy includes apparently well-established states at 0, 111, 364, 904, 1001, and 1150 keV and, in addition, a state which Kinsey and Bartholomew[43] suggest may be present at 690 keV. All of our spectra could be fitted satisfactorily in terms of the well-established transitions alone. The intensities required to give the best fits are listed in Table III. It is seen that the transition to the 690-keV state, if present at all, is too weak in all spectra to influence the values of A_0 and A_1.

[43] B. B. Kinsey and G. A. Bartholomew, Can. J. Phys. 31, 1051 (1953).

D. Se^{77} Spectra

The sample used in the study of Se^{77} was selenium metal powder about 5.0 g/cm² thick. Both the singles and the sum-coincidence spectra were recorded. Two of the singles spectra are given in Fig. 8. One spectrum is for a resonance known to have $J=1$ and the other for a resonance with $J=0$.

When the study of the resonances of Se^{77} was started, it seemed probable that all of the resonances in selenium at a neutron energy less than about 1 keV had been detected in the measurements of LeBlanc *et al.*[44] It was soon found, however, that the detection of capture gamma rays revealed many resonances which had been missed in the transmission experiments. It was, therefore, necessary to undertake a thorough investigation of the resonance of selenium. The results obtained are being reported elsewhere.[45] As is demonstrated there, Se^{77} has resonances at 113, 290, 340, and 475 eV with $J=1$ and resonances at 209 and 687 eV with $J=0$. In addition to these relatively strong resonances, numerous very weak resonances were also detected. Even though some of these are known to be in Se^{77}, their spectra are not used in the present paper—the statistical accuracy is too poor. It is felt that this selection process does not introduce a bias in the study of the partial radiation widths, since the selection depends only on the *neutron* width of a resonance.

The analysis of the Se^{77} spectra is entirely straightforward. The high-energy portion of each Se^{77} spectrum was treated under the assumption that it is formed by transitions to the known states in Se^{78} at 0, 615, and 1307 keV (Fig. 5). The basic line shape for these transitions was determined from the 9.30-MeV line in the thermal capture gamma-ray spectrum of iron and from the intense line at 9.88 MeV in the spectrum of the 340-eV resonance of Se^{77}. The correction for sum pulses, a large correction for some of these spectra, was made with accuracy from sum-coincidence data. The intensities obtained from the spectral analyses are given in Table IV.

[44] J. M. LeBlanc, R. E. Coté, and L. M. Bollinger, Nucl. Phys. 14, 120 (1959).

[45] R. E. Coté, L. M. Bollinger, and G. E. Thomas (to be published).

TABLE IV. Intensities of high-energy transitions to low-energy states in Se^{78}. The errors listed are standard statistical uncertainties.

Resonance energy (eV)	Energy of final state (keV) 0	615	1307	$[A_0+(E_0/E_1)^3A_1]$
113	36±14	38±22	319±48	81
290	142±25	552±48	−50±68	891
340	83±14	2281±40	−68±55	2808
475	376±82	1129±153	20±162	1725
Mean	159	1000	55	

V. EXAMINATION OF THE LINE SHAPE

The meaningfulness of the weak intensities presented in the previous section depends to an important degree on the accuracy of the line shape used in the analysis. Thus, it seems wise to attempt to set quantitative limits on the magnitude of the errors in intensity that might be introduced by possible errors in the line shape. The accuracy of the line shape ordinarily is important only when a weak line is somewhat obscured by a strong one. When the weak line is at a higher energy than the strong one, an error can be introduced only if the shape or width of the Gaussian-like full-energy peak of the reference line is wrong. For the W^{183}, Pt^{195}, and Hg^{199} data, it is easy to check on the possibility of an error of this kind by comparing the full-energy peak of the reference line with full-energy peaks of complex spectra that are dominated by the ground-state transition. Favorable cases for such a comparison are the 7-eV resonance of W^{183}, the 12-eV resonance of Pt^{195}, and the 34-eV resonance of Hg^{199}. In no case does the shape of the full-energy peak of these spectra show a statistically significant difference from the line shape used to analyze it. Thus, in such cases as the 175-eV resonance of Hg^{199}, for which the ratio a_0/a_1 is small, we can be sure that errors in the reference line shape used for the strong transition to the first excited state do not introduce a statistically significant error in the intensity of the weak ground-state transition. Indeed, for all of the spectra studied, errors of this kind are negligibly small since in every case they are much less than 1% of the strong transition.

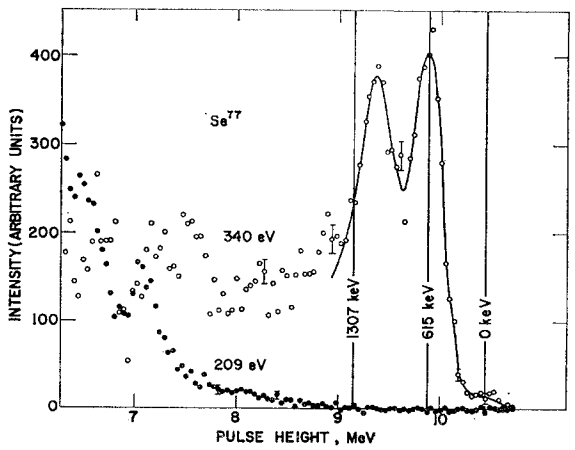

FIG. 8. Pulse-height spectra for resonances of Se^{77}. One spectrum is for a resonance with $J=1$ (340 eV) and the other is for $J=0$ (209 eV). Notice the striking difference in shape. The solid curve through the data points for the 340-eV resonance shows the least-squares fits made to obtain A_0, A_1, and A_2.

When we turn to the case in which the weak transition is at a lower energy than the strong one, it is more difficult to determine limits of error. As mentioned in Sec. II, the principal known uncertainty in the reference line shape used for W^{183}, Pt^{195}, and Hg^{199} results from an uncertainty of about ±3% in the relative intensity of the iron line 365 keV below the main line. Since the energies of the first excited states of Pt^{196} and Hg^{200} are both at roughly this energy, the principal influence of an error in the intensities of the lines in the iron spectrum is to introduce a systematic error in the values of a_1 for the resonances of Pt^{195} and Hg^{199}; the value of a_2 for Pt^{195} would be relatively free of error. Fortunately, the magnitude of this kind of possible error in a_1 can be measured by comparing the reference line shape with the 7.5-eV spectrum of W^{188}. Over a fairly wide range of energy this spectrum has significant contributions from only two transitions (a_0 and a_1) and the uncertainty in the intensities of these transitions is not enough to make a significant difference in the *shape* of a line derived from the complex spectrum. A quantitative comparison of the shape derived from the W^{183} spectrum with the reference line shape that depends primarily on the iron spectrum indicates that the systematic error in small values of a_1/a_0 is -0.008 ± 0.020 of the value. That is, there is no evidence that there is a systematic error; but the statistical uncertainties are such that the systematic error in a_1 could be ±2% of a_0. Unfortunately, this degree of uncertainty is roughly the same as that with which we began this discussion.

Since the line shape used for the Se^{77} spectra was partially derived from one of the complex spectra, a completely objective assessment of its accuracy is difficult. However, in that both a_0/a_1 and a_2/a_1 are small for the resonance (340 eV) from which the shape was derived and since the low-energy states of Se^{78} are widely separated, it is clear that the errors in intensity that result from errors in line shape cannot be large. It is estimated that the systematic uncertainty of this kind is $\Delta a_0=\pm0.005a_1$ when $a_0\ll a_1$ and $\Delta a_1=\pm0.05a_0$ when $a_1\ll a_0$. Note that no ground-state line in any spectrum studied is strong enough that its shape could make a significant error in a_1.

VI. A MONTE CARLO METHOD OF STATISTICAL ANALYSIS

The problem of deducing the shape of the distribution of partial radiation widths from the small number of widths we have measured is similar to that faced by Porter and Thomas[3] in attempting to determine the

nature of the distribution of reduced neutron widths. In both cases the body of data to be treated consists of several independent statistical samples that are all thought to be governed by distributions having the same general shape, even though the several sub-samples have unknown and different mean values. As mentioned earlier, in treating the neutron data is was assumed that the widths are distributed in a χ^2 distribution with ν degrees of freedom [Eq. (1)]. The parameter ν and the uncertainty associated with it were determined by the method of maximum likelihood,[46] a method which permits one to take account of the effect of one's ignorance about the true mean value of the populations from which the sub-samples are drawn.

In treating the partial radiation widths, we follow Porter and Thomas in assuming that the experimental widths are a statistical sample drawn from populations that are governed by a χ^2 distribution. However, two characteristics of the gamma-ray data suggest the desirability of using a different statistical treatment to find ν. First, the small size of our samples makes it probable that the maximum-likelihood estimator of ν is governed by a distribution that is highly skewed when ν is small. Thus, it is probable that the customary formulas for determining the estimator and the variance of the estimator would give results of uncertain meaning for our small sample. Second, as is emphasized throughout this paper, experimental limitations preclude intensity measurements of very weak transitions; at most, one can only set an upper limit to the intensity. The statistical analysis should take account of the effect of this experimental bias.

In an effort to obtain the maximum amount of information from our small statistical samples and to correct for the large uncertainties in the values of the small widths, we have devised a method of hypothesis testing that uses the maximum-likelihood method only as a tool for comparing different samples. The general idea[47] of the method is to compare the *physical* sample obtained in the experiment with a set of equivalent *mathematical* samples that are drawn from populations for which the value of ν is known.

First, consider the case in which there is no experimental limitation of measurement, i.e., only the small size of the statistical sample causes difficulty. The mathematical samples are formed by a Monte Carlo calculation in which widths are randomly drawn from a population governed by a χ^2 distribution for which ν has a known value ν_0. Each mathematical sample is generated in such a way that it consists of several sub-samples with independent mean values exactly as the physical sample does. Thus, there is no difference in the nature of the physical and the mathematical samples except that the former is drawn from distributions governed by an unknown value of ν, whereas the latter is drawn from distributions for which ν has a known value ν_0. The hypothesis that the physical data are drawn from a distribution having the assumed value ν_0 may, therefore, be tested by comparing the physical and mathematical samples. The comparison is made by comparing the maximum-likelihood value ν_p obtained from the physical sample with the distribution of maximum-likelihood values ν_m obtained from the mathematical samples. If ν_p falls near the center of the distribution of the ν_m, the physical data are consistent with the value ν_0 assumed for ν; on the other hand, if ν_p falls far out in the tails of the distribution of ν_m, then the physical data are inconsistent with ν_0.

The Monte Carlo technique of hypothesis testing is illustrated in Fig. 9 for the partial radiation widths of the Pt^{195} resonances. As given in Table II, the experimental data consist of three sub-samples, each of which has eight members. Let us assume that the mean values of the sub-samples are unknown and unrelated to each other. Then, the experimental widths give $\nu_p=2.06$. By drawing mathematical samples of the same kind from distributions having $\nu_0=1$ (but still neglecting experimental errors) we obtain the solid-line histogram given in the upper part of Fig. 9. The value $\nu_p=2.06$ is seen to be larger than most values of ν_m. Similarly, the lower part of Fig. 9 gives the distribution of ν_m for $\nu_0=3$. In this case, it is seen to be most improbable for ν_m to be as small as 2.06. Therefore, were it not for experimental errors in the widths, we could conclude that the true value of ν is in the range from 1 to 3.

The two solid-line histograms of Fig. 9 are instructive for the insight they afford concerning the use of the maximum-likelihood method with small samples. In each histogram we see that, although the mathematical samples are drawn from populations for which the true value of ν is a known value ν_0, the derived value ν_m is only infrequently as small as ν_0. Thus, the value ν_p derived from a single set of experimentally determined widths will usually be quite significantly larger than the true value of ν. This implies that the maximum-likelihood estimator of ν is not a very reliable estimator when used with a small sample drawn from a population governed by a small value of ν. This use of a biased estimator does not influence the accuracy of our result, of course, since the estimator is used only as a means of comparing samples.

When the measured values of the partial radiation widths are subject to experimental error, it is no longer feasible to maintain an accurate correspondence between the physical and mathematical samples. Thus, we have adopted procedures that are known to be only roughly valid but are thought to yield results that are at

[46] M. G. Kendall in *Advanced Theory of Statistics* (Hafner Publishing Company, Inc., New York, 1951), Vol. II, Chap. 17.

[47] The technique described here is an application of the general idea that the *mathematical* problem of obtaining information about the distribution of an estimator for a small sample can be solved by random sampling. This idea has rarely (if ever) been used in treating the data of physics, even though the idea is an old one to statistics. For example, see *"Student's" Collected Papers*, edited by E. S. Pearson and John Wishart (Cambridge University Press, Cambridge, 1942).

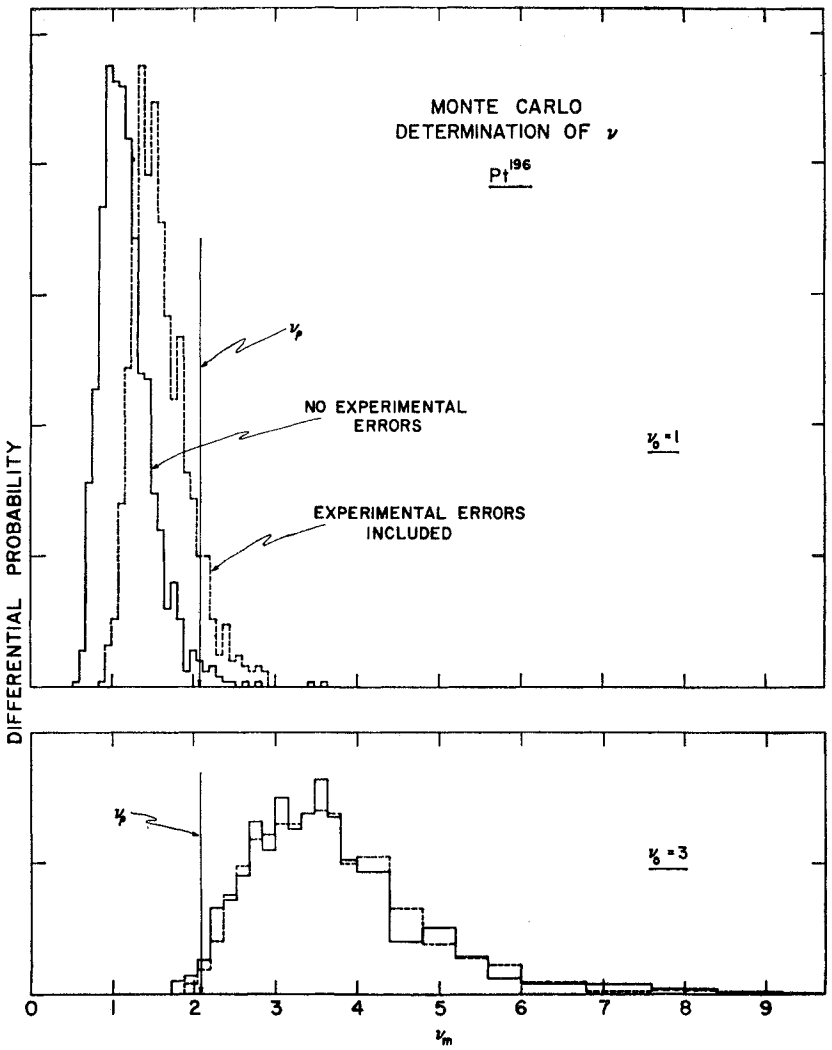

FIG. 9. Differential distribution of ν_m for mathematical samples corresponding to the Pt^{195} data.

least as accurate as is justified by the statistical size of the samples studied. In view of the order-of-magnitude fluctuations in the widths being considered, we assume that small relative errors $\Delta a_k/a_k$ in the experimental widths do not cause a significant error in the derived value of ν and, hence, can be ignored. Then, to treat the transitions for which $\Delta a_k/a_k$ is not small, we attempt to modify the physical and the mathematical width in the same way. Specifically, when $\Delta a_k/a_k<1$, we use the value a_k itself in treating the physical data to determine ν_p; and when $\Delta a_k/a_k>1$, we use the values Δa_k instead of a_k—in recognition of the fact that for a transition of this kind we have in effect measured only an upper limit to a_k. The same procedure is then used in the mathematical experiments, except that here the value of Δa_k for a particular line is an estimate of the smallest intensity that could be detected experimentally under the conditions that were present in taking the various spectra. Thus, although the data are distorted in a rather arbitrary way, at least the same kind of distortion is being applied to the physical and mathematical data.

The effect of placing lower limits on the values of the widths is demonstrated by the histograms of Fig. 9. As might have been expected, this procedure has an important influence on the derived value of ν_m if the true value is $\nu=1$. In particular, notice that it is now very unlikely to generate a value ν_m as small as 1.0. On the other hand, the influence of the limitation on observing small values of a_k rapidly becomes less important as ν_0 increases and can almost be neglected for $\nu_0=3$. Comparing the physical value ν_p with the corrected distributions of ν_m shows that whereas ν_p tended to be improbably large for $\nu_0=1$ when the experimental errors were not taken into account, the correction for the effect of these errors makes ν_p consistent with the value $\nu=1$. Similarly, it becomes even less probable that ν could be as large as 3.

To obtain information about the probable range within which the true value of ν lies, we continue in the way described above to test the hypothesis that ν_p is

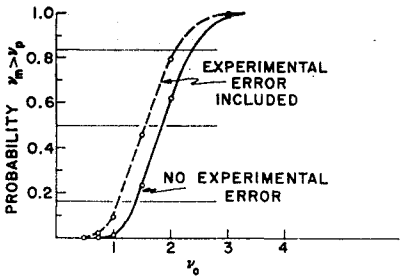

FIG. 10. Summary of the results of Monte Carlo calculations for the Pt^{195} data.

drawn from a population governed by various values of ν_0. The results obtained may be summarized in the form of the curves given in Fig. 10, in which the probability that $\nu_m > \nu_p$ is plotted as a function of ν_0. Translation of the information contained in these curves into a numerical statement must, of course, involve somewhat arbitrary definitions. However, in line with the customary usage in experimental nuclear physics, it seems reasonable to say that a physical sample corresponds to a true value $\nu \pm \Delta\nu$, where ν is defined by the statement that there is a 16.3% probability that the true value is less than $\nu - \Delta\nu$ and the same probability that the true value is greater than $\nu + \Delta\nu$; that is, $\Delta\nu$ is in the nature of a standard error for a normal error function.

In addition to the statistical uncertainty $\Delta\nu$, there is a systematic uncertainty associated with the correction for experimental errors. Let us label this systematic uncertainty $\pm\delta\nu$ and make the reasonable assumption that the magnitude of $\delta\nu$ is half of the amount by which the corrected curve of integral probability (Fig. 10) is displaced from the uncorrected curve. Then the experimental result may be summarized by writing $\nu = \bar{\nu} \pm \Delta\nu \pm \delta\nu$. For example, the data presented in the curves of Fig. 10 may be described by the result $\nu = 1.56 \pm 0.51 \pm 0.14$. Note that $\Delta\nu$ is an indication of the effective statistical *size* of the sample and $\delta\nu$ is an indication of the *quality* of the experimental data. To avoid the need to display an excessive number of curves, in the remainder of this paper the derived information about the probable range of the true value of ν will be given in the numerical form indicated above.

VII. DERIVED VALUES OF ν

The discussion in the preceding section outlined what is believed to be an accurate and objective way of obtaining quantitative information about the shapes of the distributions from which our small samples of radiation widths are drawn. These procedures are now used to treat the remaining experimental data and various combinations of the data. The derived values of ν for the various nuclides considered are summarized in Table V. In addition, Table V lists values of a quantity $P_2(\nu_m \leqslant \nu_p)$ that enables one to decide whether the experimental widths are consistent with the hypothesis $\nu = 2$. Specifically, $P_2(\nu_m \leqslant \nu_p)$ is the probability that $\nu_m \leqslant \nu_p$ for $\nu_0 = 2$ when experimental errors are included.

TABLE V. Derived values of ν. The second column, labeled n, gives the total number of widths in each sample. As explained in the text, $\Delta\nu$ is a standard statistical uncertainty dependent on the size of the statistical sample and $\delta\nu$ is a systematic uncertainty resulting from errors in measurement. The quantity $P_2(\nu_m \leq \nu_p)$ is the probability that $\nu_m \leq \nu_p$ when $\nu_0 = 2$.

Target	n	ν	$\Delta\nu$	$\delta\nu$	$P_2(\nu_m \leq \nu_p)$
Hg^{199}	6	1.24	1.2	0.12	0.28
Pt^{195}	24	1.56	0.51	0.14	0.20
W^{183}	10	1.12	0.8	0.3	0.15
Se^{77}	8	0.96	0.9	0.3	0.11
Mean	48	1.34	0.33	0.21	0.037
Gd^{155}	12	0.72	0.36	0.31	0.010
Yb^{173}	6	2.8	2.0	0.02	0.72
Hf^{177}	8	1.26	0.8	0.23	0.20
Hg^{201}	3	0.68	1.0	0.11	0.16
Mean	29	1.14	0.44	0.21	0.04

Some of the details involved in the statistical analysis of the widths are now considered.

A. Hg^{199}

To obtain a value of ν_p we use the widths for transitions to the ground state and first excited state for all of the resonances listed in Table I. The average values of these two classes of transitions are assumed to be different and unknown. The other transitions are not treated because of the doubtful reliability of their intensities. The only serious experimental bias that might be present in this selection of data for analysis would result from our exclusion of the weak transitions for the 130-eV resonance. Although it is highly probable that $J = 0$ for this resonance, the uncertainties in the transition probabilities are large enough to permit the assignment $J = 1$ with a probability of a few percent.[39] The derived values of ν would, of course, be smaller if the small widths associated with the 130-eV resonance were included. The value obtained when they are excluded is $\nu = 1.24 \pm 1.22 \pm 0.12$. It is noteworthy that, in spite of the small size of the sample, the data require that ν lie within a rather restricted range of values.

B. Pt^{195}

To obtain a value of ν_p we use the widths for transitions to the ground state, first excited state, and second excited state for all eight of the resonances listed in Table II, twenty-four widths in all.

Because of their favorable characteristics, the spectra of the Pt^{195} resonances have also been studied by other groups of experimenters. Although, as mentioned in the introduction, some experimenters initially came to the conclusion that the partial radiation widths exhibit very little fluctuation and, hence, that ν is a large number, there is now general agreement with our conclusion that ν is a small number. Closer examination shows, however, that there still remain quantitative discrep-

ancies between our results and those of the other measurements. Since these discrepancies might seem to cast doubt on the validity of our result, it is desirable to consider the data in some detail. First, let us compare the widths themselves. The data for the comparison are given in Table VI, which include the results of Julien

TABLE VI. Comparison of partial radiation widths for Pt^{195} resonances. The intensities given in heavy type indicate the transitions that were used to normalize the four sets of data.

E_0 (eV)	Reference[a]	A_0	A_1	A_2
12	ANL	**1000±15**	86±15	25±17
	BNL	**1000±42**	151±95	<85
	Harwell	**1000**	400±100	<20
19	ANL	184±5	91±7	96±7
	BNL	201±42	99±64	218±63
	Harwell	160±30	230±50	250±30
67.4	ANL	860±25	749±32	44±32
	BNL	645±49	754±78	92±78
	Harwell	450±80	510±100	120±30
	Saclay	883		
112	ANL	233±12	285±20	155±20
	BNL	218±35	299±60	218±63
	Saclay	0		
120	ANL	**604**±17	49±17	192±20
	BNL	522±49	105±78	63±63
	Saclay	**604**		
140	ANL	118±10	96±17	1071±32
	BNL	99±32	32±49	1000±74
	Saclay	604		
151	ANL	111±12	27±15	32±17
	BNL	104±42	99±78	186±95
	Saclay	327		
189	ANL	305±12	44±27	403±34
	BNL	359±56	151±95	345±95
	Saclay	1030		

[a] ANL, this paper; BNL, Ref. 19; Harwell, Ref. 18; Saclay, Ref. 16.

et al.,[16] Bird,[18] and Chrien *et al.*[19] The Argonne and Brookhaven[19] data are seen to be in generally good agreement. It is only for the weak transitions that numerical values differ substantially, and in every case the difference could result from the quoted experimental errors. The Harwell[18] data are also consistent with our own, insofar as the ground-state transitions are concerned, but the ratios A_1/A_0 are in serious disagreement. A comparison of the spectra presented in Ref. 18 with the corresponding spectra in Fig. 6 of the present paper leaves little doubt as to which set of gamma-ray intensities is more likely to be correct. Finally, the Saclay[16] data bear so little resemblance to our own or to the Brookhaven data that an outside observer would be justified in suggesting that different nuclides had been studied!

In view of the serious discrepancies between the widths reported in the present paper and those reported in Refs. 16 and 18, nothing is gained by comparing derived values of ν. Thus, we consider only the result of Chrien *et al.*[19] They report that $\nu=2.0_{-0.4}^{+0.8}$. Although this result might be said to be in satisfactory agreement with our own result $\nu=1.56\pm0.51\pm0.14$, it is in disagreement in the sense that it appears to be inconsistent with the value $\nu=1$ whereas our result is not. In view of the generally satisfactory agreement between the experimental widths themselves, it seems probable that this difference in the derived values of ν is caused by some of the systematic effects mentioned in Sec. I. In particular, one suspects that the statistical analysis used in Ref. 19 gives a result that is biased by the small size of the sample (36 widths[39]) and by the very large errors associated with the many small widths. To check this possibility, the Brookhaven data have been analyzed with our Monte Carlo procedure. The result obtained is $\nu=1.65\pm0.48\pm0.32$. This value is in very good agreement with our own and it is now not inconsistent with the value $\nu=1$. One sees, then, that the recent suggestion by Porter[48] that some special mechanism is present in high-energy radiative transitions is not a necessary conclusion from the data cited by him.

C. W^{183}

To obtain a value of ν_p we use the widths for transitions to the ground state and first excited state for all of the resonances listed in Table III. The result obtained is $\nu=1.1\pm0.8\pm0.3$. Since the intensity of individual transitions has not been measured in other experiments, this result cannot be compared with others.

D. Se^{77}

To determine ν_p we use the intensities of the two transitions of highest energy in the spectra of the resonances at 113, 290, 340, and 485 eV. The transitions to the second excited state are not used because of the possibility that transitions to neighboring states at higher energy influence the derived values of A_2. In view of the large errors in the measured intensities for the 113-eV resonance, one is tempted to ignore its intensities. However, since it seems almost certain that $J=1$ for this resonance[45] and since the relative errors $\Delta A/\langle A\rangle$ are not large, a failure to use the data would clearly introduce a bias in favor of large ν_p.

The statistical analysis of the radiation widths for the resonances of Se^{77} gives the result $\nu=1.0\pm0.8\pm0.3$. No other quantitative information is available for comparison with this result.

E. Gd^{155}, Yb^{173}, Hf^{177}, and Hg^{201}

In a study of the behavior of the *average* values of partial radiation widths, Carpenter[15] has observed the gamma-ray spectra resulting from neutron capture in resonances of Nd^{143}, Nd^{145}, Sm^{147}, Sm^{149}, Gd^{155}, Yb^{171}, Yb^{173}, Hf^{177}, and Hg^{201}. These spectra are similar to those studied in the present experiment but various experimental factors make them somewhat less suitable

[48] C. E. Porter, Nucl. Phys. **40**, 167 (1962); Phys. Today **16**, 26 (1963).

for a study of the distribution of partial radiation widths; that is, one fears that the distributions obtained from the measurements may be distorted by experimental bias. Nevertheless, it is felt that reliable information can be obtained by carefully selecting and using only those data that are most free from uncertainty.

The primary ways in which a distribution of widths obtained from Carpenter's data might be biased stemmed from our ignorance about the J values of most of the resonances and from the criteria used in selecting resonances for study. The latter effect is believed to be unimportant, even though the complexity of some of the time-of-flight spectra made it necessary to reject numerous resonances from consideration. The acceptance of a resonance for study ordinarily depended primarily on its being well enough resolved to permit the determination of a normalizing factor that is proportional to the number of captured neutrons. Because of the techniques used to determine the normalizing factor, the criterion of acceptance depended strongly on the neutron width and on the total radiation width but was largely independent of partial radiation widths.

The second point of uncertainty, our ignorance about the J values of the resonances, is clearly more serious since the mean value of a particular class of transitions depends on its J value. However, one may be able to find cases in which the uncertainty in the mean values is unimportant. For example, consider the 7.3-MeV transitions to the 4^+ state of Hf^{178} from the 3^- and 4^- states formed by capture in Hf^{177}. If the mean values for transitions from these two kinds of initial states are proportional to the average level spacings[15,49] and if the level spacing[1,2] is proportional to $(2J+1)^{-1}$, as the existing evidence indicates, the mean values of intensities would be in the ratio 9/7. This ratio is so near to unity that it seems reasonable to suppose that the distribution formed by all 7.3-MeV transitions, independent of the initial J value, would be nearly indistinguishable in *shape* from the distribution for the transitions from states with a specified value of J. Numerical calculations show that this hope is fulfilled when ν is a small number and when the ratio of the mean values of widths is not more than 2. Thus, although recognizing that the ratio of mean widths cannot be known with certainty, we select for statistical analysis those data of good quality for which the expected ratio of mean widths associated with the two initial states is close to unity.

The transitions that satisfy our selection criteria are the following: $(1^-,2^-) \rightarrow 2^+$ in Gd^{156}, $(2^-,3^-) \rightarrow 2^+$ in Yb^{174}, $(3^-,4^-) \rightarrow 4^+$ in Hf^{178}, and $(1^-,2^-) \rightarrow 2^+$ in Hg^{202}. For each kind of transition we use all of the widths listed in Ref. 15. The values of ν obtained from the statistical analysis of these data are summarized in Table V.

[49] J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley & Sons, Inc., New York, 1952), Chap. 12.

F. Mean Values of ν

If one makes the physical assumption that the basic shape of the distribution of widths is the same for all nuclides of a given class, then for these nuclides one can obtain a weighted mean value of ν by a straightforward application of the statistical technique outlined in Sec. VI. Average values of ν have been derived in this way for the two groups of nuclides treated in this paper. The over-all statistical sample used in each calculation was formed from all of the widths used to obtain values of ν for the individual nuclides. For the transitions in Se^{78}, W^{184}, Pt^{196}, and Hg^{200}, the over-all result is $\nu=1.34\pm0.33\pm0.21$. For the other group of nuclides (Gd^{156}, Yb^{174}, Hf^{178}, and Hg^{202}), the over-all result is $\nu=1.14\pm0.44\pm0.21$.

VIII. CORRELATIONS

Several papers have suggested that the partial radiation widths might exhibit correlation effects. On theoretical grounds, Porter and Thomas[3] felt that there might be a *positive* correlation between the widths for radiative transitions from some single high-energy state to several low-energy states of the same character. The Brookhaven group,[2,13,50] on the other hand, have expressed the view that the experimental data indicate the existence of a *negative* correlation or anticorrelation, between widths of this kind. [The implications for the statistical theory of negative correlations has recently been considered by N. Rosenzweig, Phys. Letters **6**, 123 (1963).] Finally, Lane and Lynn[51] have proposed a model of radiative transitions that would in some cases require the partial radiation width to be positively correlated with the reduced neutron width. It clearly is important for us to examine the available data for these possible effects since the presence of any any of them to a significant degree might prejudice the validity of the results on the distribution of partial radiation widths.

A. Correlation between Partial Radiation Widths

One way of searching for the existence of correlation between the widths for radiative transitions to several low-energy states is to examine the distribution of the *sum* of the widths. If the distribution of widths for individual transitions is a χ^2 distribution with ν degrees of freedom and if the individual transitions are uncorrelated, then the distribution of the sum of widths to n final states would be expected to be a χ^2 distribution with $n\nu$ degrees of freedom. On the other hand, a positive correlation would result in a distribution with less than $n\nu$ degrees of freedom and a negative correlation would result in a distribution with more than $n\nu$ degrees of freedom. These arguments were first applied

[50] R. E. Chrien, H. H. Bolotin, and H. Palevsky, Bull. Am. Phys. Soc. **6**, 69 (1961).
[51] A. M. Lane and J. E. Lynn, Nucl. Phys. **17**, 563 (1960).

to the partial radiation widths by Hughes.[13] He pointed out that although we had reported[7] that the partial widths for transitions to the individual low-energy states of W^{184} showed large fluctuations, the sum of widths for transitions to the ground state and first excited state was roughly constant.[6] Similarly, Chrien *et al.*[50] at one time reported that the distribution of the sum of transitions to the three lowest states of Pt^{196} was narrower than expected on the basis of a Porter-Thomas distribution for transitions to the individual states. If statistically meaningful, these observations reveal the presence of negative correlation effects.

Let us use the approach described above to examine the widths reported in the present paper. Consider first the data for the resonances of Pt^{195}. We start by forming the sum $\sum_j A_j E_j^{-3}$, where E_j is the gamma-ray energy; implicit in this form of the sum is the assumption that the mean values of the partial radiation widths are proportional[49] to E_j^3. One may now treat the statistical sample of sums by means of the Monte Carlo calculation described in Sec. VI. If the data for the sum of widths of all eight resonances listed in Table II are used, we obtained $\nu_p=5.4$. There is a 24% probability for ν_m to be as large as 5.4 when $\nu_0=3$; hence, there is no indication that the transitions to the three lower states of Pt^{196} are not independent. This finding agrees with the final conclusion of the Brookhaven group.[19]

The same treatment outlined above can also be applied to the sum of widths for the resonances of W^{183}, the other case which has been cited as giving evidence for a negative correlation between transitions. If we use the data given in Table III for the five resonances studied in the present experiment, we obtain the value $\nu_p=11.5$. Comparing this value with the ν_m distribution for $\nu_0=2$, one finds that ν_p is much greater than most values of ν_m, in qualitative agreement with the assertion that the sum of widths is surprisingly uniform. However, if the matter is considered quantitatively, the distribution of ν_m is found to be so broad that ν_m is greater than ν_p for 5.6% of the mathematical samples. Thus, the apparent uniformity of the sum of widths for the first five resonances of W^{183} could occur by chance and should not be considered to be more than an indication of a need for measurements on a greater number of resonances. Measurements on other resonances at higher energy have been reported in Ref. 16. However, in view of the serious discrepancies between the data of Ref. 16 and our own data for both W^{183} and Pt^{195} and since no evidence is presented in Ref. 16 to indicate that weak radiative transitions, if present, could have been detected, it seems best to restrict the present discussion to our own data.

Although an examination of the distribution of the sum of widths is a worthwhile way to search for correlation effects, its usefulness depends on the assumption that the distribution of widths for individual transitions is known. Moreover, it seems probable that an examination of the distribution of the sum is not a very sensitive way to search for correlation. Let us attempt, then, a direct test of the hypothesis that the two samples formed by transitions from a set of initial states to two final states are independent. We may test this hypothesis by seeing whether or not the correlation coefficient for the two samples differs from zero to a statistically significant degree.[52] If the widths in one sample are labeled α_i and the widths in the second sample are labeled β_i, where i is the label on the initial state, the correlation coefficient for the widths is defined as

$$r_{\alpha\beta}=\frac{\sum_i(\alpha_i-\bar{\alpha})(\beta_i-\bar{\beta})}{[\sum_i(\alpha_i-\bar{\alpha})^2\sum_i(\beta_i-\bar{\beta})^2]^{1/2}},$$

where $\bar{\alpha}$ and $\bar{\beta}$ are the mean values of the two samples.

Let us first treat the tungsten data by comparing the intensities of transitions to the ground state and first excited state of W^{184}. The data of Table III yield the result $r_{01}=-0.65$ for the correlation coefficient. The question whether or not this value is a statistically significant indication of a negative correlation may be answered by examining the distribution of the estimator r for samples that are known to be independent. It is known to be generally true,[52] even for small samples and independent of the form of the distribution of α and β, that the distribution of r is approximately of the form $(1-r^2)^{n/2-2}$, where n is the number of elements in each sample. Thus, for the tungsten data the distribution is of the form $(1-r^2)^{1/2}$. This implies that a wide range of values of r are almost equally probable. It is immediately apparent that the value $r_{01}=-0.65$ is entirely consistent with values that would be expected if the transitions to the two states of lowest energy in W^{184} are independent. Thus, the correlation test gives no indication that the widths are not independent.

The above test for independence is undoubtedly valid; but it may not be very sensitive since, in the absence of a physical theory indicating how the transitions might fail to be independent, there is no reason to suppose that there would be a linear regression between the two sets of *widths*. There could equally well be a linear regression between the two sets of values of any *function* of the widths. Thus, to make the test for independence more comprehensive, we calculate the correlation coefficient for various functions of the widths. The functions considered are x^2, $(1+x)^2$, $\ln x$, and $x^{1/2}$, where $x=\Gamma/\bar{\Gamma}$. The results obtained are given in Table VII. In no case is the absolute value of r large enough to be inconsistent with independence of the samples.

The procedure outlined above may also be used to compare the transitions to the two lowest states of Se^{78} and the three lowest states of Pt^{196}. The results obtained for the correlation coefficients are given in Table VII.

[52] M. G. Kendall and A. Stuart, *Advanced Theory of Statistics* (Charles Griffin and Company Ltd., London, 1961), Vol. 2, Chap. 22.

TABLE VII. Correlation coefficients $r_{\alpha\beta}$ for various functions of $x \equiv \Gamma/\bar{\Gamma}$. The subscripts 0, 1, and 2 on r refer to the final states in radiative transitions. The subscripts n and γ designate reduced neutron widths and the sum $\Sigma_j A_j E_j^{-3}$, respectively. The number of elements in the samples being compared is 9 for Pt^{195}, 5 for W^{183}, and 4 for Se^{77}.

Target	$r_{\alpha\beta}$	x	$\ln x$	$x^{1/2}$	x^2	$(1+x)^2$
Pt^{195}	r_{01}	+0.44	+0.42	+0.43	+0.45	+0.45
	r_{12}	−0.24	−0.14	−0.23	−0.18	−0.21
	r_{20}	−0.44	−0.45	−0.48	−0.32	−0.38
	$r_{n\gamma}$	−0.19	−0.15	−0.16	−0.27	−0.24
W^{183}	r_{01}	−0.65	−0.67	−0.68	−0.53	−0.60
	$r_{n\gamma}$	−0.03	−0.02	−0.02	−0.03	−0.03
Se^{77}	r_{01}	+0.13	+0.66	+0.38	−0.14	−0.06
	$r_{n\gamma}$	+0.84	+0.81	+0.79	+0.94	+0.90

In no case is there a statistically significant indication of correlation between the widths.

B. Correlation between Radiation and Neutron Widths

A qualitative examination of the thermal-neutron-capture gamma-ray spectra seems to show that some particular transitions are so strong that a special mechanism is required to account for them. Lane and Lynn[51] have suggested that this mechanism consists of a direct transition by the incident s-wave neutron into low-energy states that are dominated by a p-wave orbital component. One implication of this model is that the partial width of a direct radiative transition would be positively correlated with the reduced neutron width Γ_n^0 of the resonance. Although the low-energy states with which our data are concerned are not expected to be of the kind for which the direct transition would be an important effect, it seems worthwhile to make the correlation calculation since the mathematical groundwork has already been laid.

The test for a correlation between the reduced neutron width and the partial radiation width was made by calculating the coefficient of correlation between the various samples of Γ_n^0 and $\sum_j A_j E_j^{-3}$, where E_j is the energy of a radiative transition to a state j. For Se^{78}, W^{184}, and Pt^{196} the sum was over 2, 2, and 3 final states, respectively. The calculated values of the coefficient are given in Table VII. Of these results, only the value $r \approx 0.9$ for the resonances of Se^{77} is suggestive of a positive effect. However, since the distribution of r for this case with $n=4$ is constant, i.e., all values in the range $-1 \leqslant r \leqslant +1$ are equally probable, the experimental data are entirely consistent with the hypothesis that the reduced neutron width and the partial radiation widths are independent.

IX. SUMMARY

As was outlined in Sec. I, the statistical assumptions that lead to the Porter-Thomas distribution for the reduced neutron width appear to apply with an equal degree of validity to the partial radiation width. All of the experimental results presented in this paper are consistent with this expectation. Moreover, the mean values of ν and the values of $P_2(\nu_m \lesssim \nu_p)$ for both of the groups of nuclides considered in Table V are small enough to make it most improbable that ν could be 2 or larger. However, since ν might not be an integer and since any value other than $\nu=1$ would be at variance with a purely statistical description of radiative transitions, it is of interest to examine other kinds of data to see if the upper limit on ν can be reduced.

One of the most reliable pieces of evidence concerning the nature of the high-energy radiative transitions is provided by the energy dependence of the cross section for a single high-energy transition. If a transition of this kind can be thought of as proceeding by way of a single exit channel, the cross section in the neighborhood of closely spaced resonances would be expected to exhibit interference effects similar to those observed in resonance scattering of neutrons. In the only experiment of this kind to be reported to date, Coté and Bollinger[42] examined the energy dependence of the intensity of the ground-state transition resulting from neutron capture in the neighborhood of two resonances of Pt^{195}. The cross section was observed to exhibit interference effects that are in quantitative agreement with the assumption of a single-channel process and in definite disagreement with the assumption of the presence of many channels. However, one cannot exclude the possibility that the experimental result could occur by chance for a reaction that can proceed by some small number of channels greater than unity.

Another source of direct information about the distribution of radiation widths is provided by the (p,γ) measurements by Carver and Jones,[53] who studied proton capture in Ni^{58}. The experiments on proton capture have an advantage over those on neutron capture in that they give information about a greater number of widths but they suffer from the disadvantage of being unable to separate $M1$ from $E1$ transitions with certainty. The experimental distribution of widths was found to be consistent with the assumption $\nu=1$ and to be inconsistent with the assumption of a large value of ν. The upper limit of ν that would be consistent with the data is not stated quantitatively. However, in view of the systematic uncertainty concerning the multipolarity of the transitions being observed, it seems improbable that these data would place a smaller upper limit on ν than is set by the data from neutron capture.

Somewhat less direct information about the distribution of widths is obtained by studying the way in which the gamma rays from capture of thermal neutrons deviate in intensity from the expected mean value for the transition. Data of this kind are free of most of the experimental uncertainties that beset the data for resonant capture but, on the other hand, the thermal

[53] J. H. Carver and G. A. Jones, Phys. Rev. Letters 3, 559 (1959).

data contain serious uncertainties of their own. The estimate of the mean value of a radiation width is necessarily rather crude, one cannot be sure of the number of resonances contributing to the thermal capture, and interference between the amplitudes for several resonances can have an important effect on the gamma-ray intensity. Bartholomew[54] has examined the thermal data and finds that they are consistent with the value $\nu=1$ and are inconsistent with a large value of ν.

Even less direct evidence concerning the distribution of partial radiation widths is that provided by the results of Carpenter and Bollinger[35] on the *total* radiation width of the resonances of Hg^{201}. For this particular nuclide, the total radiation widths were found to fluctuate much more than is observed for most of the cases that have been studied. Although a part of the variation can be accounted for by a possible difference in the spins of the several resonances that are studied, a quantitative explanation of all of the observed widths requires another source of fluctuation such as would be provided by a distribution of partial radiation widths that is characterized by a small value of ν.

The above outline shows that there is by now a substantial body of evidence indicating that the distribution of partial radiation widths is extremely broad. In contrast, in the opinion of the authors, there is only one kind of experimental evidence that needs to be considered as a significant indication that the distribution might be rather narrow for some kinds of transitions. These data are those from measurements on the high-energy gamma rays from resonant capture of neutrons in U^{238}. In these measurements, which were first undertaken with the Brookhaven-Chalk River chopper,[20] the relative intensity of pulses with heights in the neighborhood of 4.02 MeV have been studied. Originally these pulses were thought to originate from a single transition to an excited state of U^{239} at about 0.60 MeV. More recently, however, it has been shown by Jackson and Bollinger[21] that at least three transitions contribute strongly to the observed counting rate. Fiebiger[55] now finds evidence for four transitions. Thus, the spectrum being considered is much more complex and poorly understood than are those treated in the present paper. Nevertheless, the reported uniformity in the intensity of the high-energy gamma rays of U^{239} cannot reasonably be explained entirely in terms of a high multiplicity of transitions, since some of the data[20,22] imply that the distribution of widths has a value of ν in the neighborhood of 50 or 100.

In the opinion of the authors, however, it would be premature to accept the neutron resonance-capture data of U^{238} as definite evidence for a deviation from the Porter-Thomas distribution for some transitions and especially not for transitions of the kind studied in the present paper. [*Note added in proof.* This conclusion is reinforced by recent experimental evidence reported by H. E. Jackson, Bull. Am. Phys. Soc. 8, 513 (1963). From refined spectral measurements at 12 resonances of U^{238} he finds that the distribution of high-energy transitions in U^{239} is characterized by $\nu=8.3\pm2.6$, a value that is not inconsistent with the expected value of about 5.] Our reservations stem from doubts about the adequacy of the gamma-ray spectrometers used in the two more extensive studies of the very complex spectra resulting from neutron capture in the resonances of U^{238}. These latter doubts are reinforced by a comparison between the three reported sets of data,[20–22] which are found to disagree in two respects. First, in the four resonances studied with the equipment described in the present paper, the degree of variation in the relative intensity of the high-energy gamma rays was found[21] to be almost twice as great as that reported by the Brookhaven group[20] for the same resonances. Second, although the Saclay group[22] report their data to be in good agreement with that of the Brookhaven group,[20] the two sets of numerical values actually exhibit a *negative* correlation that cannot reasonably be explained by the reported experimental errors.

In summary, the data now available show that the distribution of widths for a wide class of high-energy radiative transitions is broad enough that the data can be well described by a χ^2 distribution with $\nu=1$, the only distribution that has any published theoretical support. However, the data do not yet permit one to exclude a few percent possibility that ν is as large as two, a result that would be of some interest because it would imply a failure in the Porter-Thomas description of high-energy transitions. Also, one cannot exclude the possibility that there is more than one class of transitions and that for at least one class the distribution of widths is so narrow as to require a large value of ν for its description.

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APPENDIX: MISCELLANEOUS NUCLEAR DATA

A. Mercury

In obtaining a value of ν for the Hg^{199} data, the 130-eV resonance was ignored under the assumption that its spin is $J=0$. This assignment is highly probable but not conclusive. There are two significant bits of information. First, the previously reported[33] relative intensity of two-step cascades to the ground state seemed to indicate that $J=0$. However, it was later

[54] G. A. Bartholomew in *Proceedings of the Conference on Electromagnetic Lifetimes and Properties of Nuclear States, Gatlinburg, Tennessee, 1961*, Nuclear Science Series Report No. 37 (National Academy of Science–National Research Council, Publication 974, Washington 25, D. C. 1962), p. 209.

[55] N. F. Fiebiger, Bull. Am. Phys. Soc. **7**, 11 (1962).

found that the observed effect might possibly result from statistical fluctuations in the intensities of individual transitions. The second source of information about the J value is the observed intensity of high-energy transitions. After correcting for the influence of sum pulses, the intensities are found to be

$$(A_0)_{130}=(-0.02\pm0.03)(A_0)_{34},$$
$$(A_0+A_1)_{130}=(-0.03\pm0.66)(A_0)_{34}.$$

Thus, the measured intensities are effectively zero but the statistical uncertainties are such that there is a probability of about 5% that $J=1$ for the 130-eV resonance.

The observed intensities of the high-energy transitions in the Hg^{199} spectra give no new information about the character of the low-energy states of Hg^{200}, but the intensities do tend to support the previous assignments. In particular, the nonzero intensities of transitions to the states at 1050 and 1267 keV are consistent with the recent assignment[41] of 0^+ and 2^+, respectively, for these states.

The small uncertainty ($\approx$2 keV) with which the least-squares fitting procedure locates the position of an isolated high-energy gamma-ray line suggests that the resonant-capture gamma-ray spectra, even if observed with a NaI scintillation spectrometer, might be a useful source of information about neutron binding energies. To test this idea, an effort was made to measure the difference in the binding energy B of Hg^{199} and Pt^{195} by simultaneously observing the spectra from capture in the 34-eV resonance of Hg^{199} and the 12-eV resonance of Pt^{195}. A composite target was used in the measurement. A least-squares matching of the full-energy peaks of the ground-state transitions of the two spectra yields the result $B_{199}-B_{195}=109\pm10$ keV. This difference is in good agreement with the value 117 keV obtained from mass values[56] and with 111 keV from thermal-capture gamma-ray energies.[30,32] The small error associated with our value, although based solely on statistical uncertainties, is believed to be realistic since the spectra being compared were similar in shape, were required under roughly equal instantaneous counting rates, and were recorded simultaneously in a single instrument. Indeed, it should be easy to measure the difference in binding energy with much smaller uncertainty than ±10 keV since, for the spectra used, the pulse height drifted badly during the course of the measurement.

B. Platinum

The spin assignment of the 155-eV resonances has long been in dispute, with Refs. 19 and 37 giving $J=1$ and Refs. 16 and 17 giving $J=0$ for the resonance. To obtain an unambiguous result, we have examined the shape of the total cross section of platinum in the neighborhood of the 151 and 155-eV resonances. These cross sections were obtained from transmission measurements on thick samples of platinum metal with an over-all resolution width of about 14 nsec/m.

The shape of the cross section of platinum between the 151 and 155-eV resonances depends sensitively on the spins of the resonances because the neutron widths are large enough to cause strong interference effects if the spins of the two resonances are the same. These interference effects are not observed. Thus, the spins of the two resonances are unquestionably different. Moreover, the shapes of the resonances indicate clearly that the 155-eV resonance is the one with $J=0$.

The assignment of $J=0$ for the 155-eV resonance causes some difficulty in the interpretation of the gamma-ray spectrum for this resonance. As may be seen in Fig. 6, there appears to be a nonzero intensity for the ground-state transition observed at this resonance, a result that implies $J=1$. A possible explanation of the observed intensity is that it results from sum pulses; however, a quantitative treatment shows that the data are inconsistent with this hypothesis. A second possibility is that multiple scattering in the relatively thick sample used in the measurement may be severe enough that most of the events observed at the time-of-flight channel corresponding to the 155-eV resonance actually result from capture in the 151-eV resonance. The usual degree of similarity in the over-all shapes of the two spectra (Fig. 6) suggests that this interpretation is correct.

The gamma-ray spectra for Pt^{195} resonances at energies greater than 190 eV were not studied by us because of the fear that single resonances could not be isolated with the available resolution. The measurements of Julien *et al.*[37] indicate that our caution was justified. They report resonances in Pt^{195} at 222, 253, 261, 278, 284, 302, and 308 eV. The pairs of resonances (253, 261), (278, 284), and (302, 308) eV could not have been resolved in either our work or that reported by Chrien *et al.*[19]

C. Tungsten

Various conflicting evidence has been reported concerning the number of resonances in tungsten in the neighborhood of 20 eV. The γ-ray measurements on tungsten help to clarify the matter. The shapes of the time-of-flight spectra are complex enough in this region to suggest the presence of at least four resonances. However, a careful examination of the γ-ray spectra shows that there is evidence for only three resonances—the well-known resonances at 18.8 and 21.2 eV and a much less prominent one at 15.9 eV. Hence, the unusual shapes of the time-of-flight spectra presumably result from multiple scattering.

The sum-coincidence data that were used to correct for summing in a single crystal may also be used in the manner described by Jackson and Bollinger[36] to give

[56] F. Everling, L. A. König, J. H. E. Mattauch, and A. H. Wapstra, Nucl. Phys. 18, 529 (1960).

information about binding energies and to make isotopic assignments of resonances. The sum-coincidence spectra yield the results 6.16±0.05 and 5.44±0.09 MeV for the binding energies of W^{182} (21.2 eV) and W^{186} (18.8 eV), respectively; these values are not in disagreement with the values 6.29±0.04 and ≈4.8 MeV obtained in other ways.[40] The maximum energy of the sum pulses observed in the spectrum for the 15.9-eV resonance is 6.30±0.08 MeV. This energy implies that the resonance must be assigned to either W^{180} or W^{182}. Similarly, a previously reported weak resonance at 75 eV must be assigned to either W^{180} or W^{182}.

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Atomic Masses from Gallium to Molybdenum*

RICHARD R. RIES,† RICHARD A. DAMEROW,‡ AND WALTER H. JOHNSON, JR.
School of Physics, University of Minnesota, Minneapolis, Minnesota
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The 16-in. double-focusing mass spectrometer at the University of Minnesota has been employed to measure the atomic mass of 42 stable isotopes in the region $A=69$ to 100. The standard error associated with these results is approximately 5 parts in 10^8. Improvements in the instrument are described that result in an increase of useful resolution of a factor of 2 to 3. A set of 64 radioactive masses is calculated from the stable mass data together with β-decay energies and nuclear reaction Q values. The resultant table of masses is used to calculate total nuclear binding energies, separation energies and pairing energies for a number of nuclei in the region near $N=50$. The systematics of the separation energies display very smooth characteristics except at the shell closure. Neutron pairing energies show a marked decrease in value following the shell closure.

INTRODUCTION

THE 16-in. double-focusing mass spectrometer at the University of Minnesota has been employed in the past to measure atomic masses in several regions of the periodic table. Measurements of atomic masses for most of the stable isotopes with $A<70$ have been reported.[1–5] In addition, krypton and xenon,[6] lead and mercury[7] masses have been measured. Operational difficulties in the mass spectrometer become progressively more apparent in measurements of heavy isotopes where maximum resolution is required. These difficulties necessitated the movement and reconstruction of the instrument.

The improved instrument has been employed to measure a number of stable masses for $A>70$. Mass doublets in the region from gallium through molybdenum are reported in this paper and the following paper[8] reports results in the region from ruthenium through xenon.

The mass results have been employed to study the nuclear binding energy systematics in the region $Z=31$ to 42 and $N=36$ to 58. These data include the $N=50$ shell closure and also the possible $N=40$ and $Z=40$ subshell closure.

THE INSTRUMENT

The instrument employed for all previous measurements has been described in some detail elsewhere.[9] The ion optics of the instrument are arranged to yield first- and second-order angle focusing and first-order velocity focusing at the fixed collector slit. Doublets are measured by the peak-matching method.

One of the recurring difficulties in the original instrument was the random modulation of the ion beam resulting from building vibrations and time-varying magnetic fields from nearby ac power lines. The modulation not only limited the maximum useful resolution of the mass spectrometer, but also limited the sweep frequency that could be employed.

To remove these difficulties, the instrument was rebuilt in a ground-floor room which had lower stray magnetic fields. The instrument was mounted on a 2-ton cast-iron surface plate. The use of the surface plate as an

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† Present address: Max Planck Institut für Chemie, Mainz, Germany.
‡ Present address: Sandia Corporation, Albuquerque, New Mexico.
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