Fission of U^{238} and Pu^{240} Nuclei Excited by Inelastic Alpha-Particle Scattering*

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Angular correlations have been measured for fission fragments emitted from nuclei excited by an (α,α') reaction. The angular correlations were measured as a function of the excitation energy of the fissioning nucleus. From an analysis of the results an attempt is made to identify the vibrational bands which are present in the transition-state spectrum below the energy necessary for two quasiparticle excitations. The transition-state spectrum which is obtained from the analysis is found to be consistent with previous $Pu^{232}(d,pf)$ results.

I. INTRODUCTION

As was first pointed out by Bohr and Wheeler,[1] many of the characteristics of the fission process can be adequately described in terms of a liquid drop model of the nucleus. In this model, as a nucleus distorts from its stable shape to scission, a certain minimum energy must be supplied to provide the difference in mass (or energy) between the lowest states of the nucleus in its distorted saddle-point configuration and its stable shape. This minimum energy corresponds to the threshold energy for fission. Starting with an excitation energy equal to the fission threshold, the nucleus is constrained to pass through the lowest excitation-energy state at the saddle point. However, if the nucleus is initially excited to an excitation energy greater than the fission threshold, then there is excess energy available at the saddle point which can go into either internal excitation of the saddle-point nucleus or kinetic energy in the unbound degree of freedom leading to fission. It is expected that for the saddle-point configuration there will be available transition states corresponding to excitation of various discrete particle and collective states similar to those available in the nucleus at its stable shape. At any given excitation above the fission threshold, the system may proceed to fission through any of the appropriate transition states that correspond to internal excitations between the fission threshold and the initial excitation energy. The properties of the transition states which are expected to occur at low excitation energies have been discussed in detail by Bohr[2] and Wheeler.[3]

Studies of these transition states are of particular interest because they can yield a direct measurement of the effect of deformation on the level structure of a nuclear system. One method that is available for the study of transition states is to investigate the angular distribution of the emitted fission fragments. The angular distributions of the fragments are dependent on the values of the total angular momentum J and the projection of the total angular momentum on the symmetry axis, K. Thus, angular distribution measurements can be expected to give information on the average values of J and K for the transition states that are available at a given excitation energy. Theoretical treatments for fission fragment angular correlations have been developed using both semiclassical[2,4] and quantum-mechanical[5,6] approaches.

Recently it has been shown that it is possible to measure the angular correlations of fragments from nuclei that have been excited by a direct reaction. In this type of reaction the excitation of the fissioning nucleus is labeled by the energy of the outgoing direct particle. In addition, a direct reaction can transfer relatively large angular momenta to the residual fissioning nucleus resulting in large anisotropies in the angular correlation. For (d,pf) measurements[7] on Pu^{239} and U^{233} targets, it has been shown that under certain experimental conditions it is possible to obtain the angular correlations from distorted-wave Born approximation (DWBA) calculations and then extract from the experimental data K_0^2, the mean square value of K, as a function of the excitation energy at the saddle-point configuration. These results gave evidence for vibrational bands in the transition state spectrum of Pu^{240} and yielded a direct measure of the pairing gap at the saddle point for this nucleus.[7] In addition, the (d,pf)

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[1] N. Bohr and J. A. Wheeler, Phys. Rev. **56**, 426 (1939).
[2] A. Bohr, in *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1955* (United Nations, New York, 1956), Vol. 2, p. 151.
[3] J. A. Wheeler, in *Fast Neutron Physics*, edited by J. B. Marion and J. L. Fowler (Interscience Publishers, Inc., New York, 1963), Part II.
[4] I. Halpern and V. M. Strutinski, in *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958* (United Nations, Geneva, 1958), Vol. 15.
[5] J. J. Griffin, in *Proceedings of the International Conference on Nuclear Physics, Paris, 1958* (Dunod Cie., Paris, 1958); Phys. Rev. **116**, 107 (1959); *ibid.* **127**, 1248 (1962); in *Proceedings of the International Conference on Nuclear Structure, Kingston, 1960*, edited by D. A. Bromley and W. E. Vogt (University of Toronto Press, Toronto, Canada, 1960).
[6] V. M. Strutinskii, Zh. Eksperim. i Teor. Fiz. **30**, 606 (1956); **39**, 781 (1960) [English transls.: Soviet Phys.—JETP **3**, 638 (1956); **12**, 546 (1961)]; Nucl. Phys. **27**, 348 (1961).
[7] H. C. Britt, W. R. Gibbs, J. J. Griffin, and R. H. Stokes, Phys. Rev. Letters **11**, 343 (1963); Phys. Rev. **139**, B354 (1965).

reaction has also been used to study transition states in odd-mass fissioning nuclei.[8]

One difficulty encountered in the study of even-even fissioning nuclei, using the (d,pf) reaction, was that the random coupling of the neutron spin and the spin of the odd-mass target nucleus to the orbital angular-momentum transfer decreases the anisotropy in the angular correlation and limits the information that can be obtained in these experiments. In particular, one consequence of target spin effects is that with present experimental techniques Pu^{239} is the only target available which is useful for studying the transition-state spectra of even-even nuclei. However, in addition to the (d,pf) reaction there are several other direct reactions which could be used to study transition states in even-even nuclei. One particularly appealing reaction is inelastic alpha-particle scattering. In this case all of the relevant spins are zero and, furthermore, the angular-momentum transfers should be significantly larger than those in the (d,p) reaction. These two factors should lead to much larger angular anisotropies for $(\alpha,\alpha' f)$ reactions than were observed in (d,pf) experiments. This expectation is confirmed in a study of the $U^{238}(\alpha,\alpha' f)$ reaction[9] where anisotropies of the order of 7 are observed for fission near threshold excitation.

In this paper results are reported for measurements of detailed angular correlations for the $(\alpha,\alpha' f)$ reaction on targets of U^{238} and Pu^{240}. The data were analyzed by means of DWBA calculations in an attempt to obtain detailed information on the K values for the vibrational bands that exist in the transition state spectrum between the fission threshold and the excitation energy corresponding to the onset of two-quasiparticle excitations.

II. EXPERIMENTAL PROCEDURES

A. General

In the experiments alpha-particle beams were obtained from the 88-in. cyclotron at Berkeley. The incident-beam energies were 40.0 and 38.1 MeV for the U^{238} and Pu^{240} measurements, respectively. The effective energy resolution due to both energy spread in the incident beam and the kinematic energy spread because of the finite solid angle of the alpha detector was 0.4- and 0.5-MeV full width at half-maximum for the U^{238} and Pu^{240} measurements, respectively.

The fission detectors were phosphorus-diffused semiconductor detectors of 400 ohm-cm p-type silicon which were operated at reverse biases of 100–200 V. Detectors of two sizes were used, 1 cm$\times$1 cm square and 1 cm$\times$2 cm rectangular. They were placed approximately 1 in. from the target. For angles near the recoil angle the square detectors were used, and they subtended a total angle of approximately 22°. The rectangular detectors were used for fission angles that were nearly perpendicular to the recoil direction. Lithium-drifted silicon detectors of 1 mm thickness were used to detect the inelastic alpha particles. The alpha detector was collimated by a vertical slit placed approximately 1 in. from the target center. The collimators used were $\sim\frac{1}{2}$ in. high and widths of $\frac{1}{8}$ in. to $\frac{1}{4}$ in. were used in various runs. A 1-mil Al foil was placed in front of the alpha-particle detector to prevent fission fragments from entering this detector.

For all of the experiments the alpha-particle detector was placed at an angle of 75° with respect to the direction of the incident beam. This angle represented the maximum angle for which the inelastic cross section was large enough to allow measurements of coincident alpha-particle spectra. This large angle was advantageous for two main reasons. First, the targets contained only impurities of C^{12} and O^{16}, and at $\theta_\alpha=75°$ the elastic scattering cross section for O^{16} has a very deep minimum.[10] This meant that for $\theta_\alpha=75°$, the first significant peak in the spectrum due to the C^{12} and O^{16} impurities was the C^{12} elastic group at $E_\alpha=24$ MeV. This allowed the observation of coincident events corresponding to an excitation energy of up to $\sim$13 MeV in the fissioning nucleus without significant accidental contributions from C^{12} and O^{16} interactions. In addition, the large angle was desirable in order to minimize the Coulomb distortion effects on the angular correlations and thus facilitate the detailed analysis of the angular correlations. The angular correlations were obtained by simultaneously measuring the inelastic alpha-particle spectra in coincidence with three or four individual fission detectors placed at various angles. Multiple fission detectors were used in order to decrease the large amount of accelerator time needed to obtain reasonable statistics with the low counting rates available.

The targets used in this experiment were made by vacuum evaporation of $U^{238}O_2$ and $Pu^{240}O_2$ onto $\sim$70-μg/cm^2 carbon backings. The deposit thicknesses were 250 and 110 μg/cm^2 for the U^{238} and Pu^{240} targets, respectively. The Pu^{240} target had an isotopic composition of 98.0% Pu^{240}, 1.19% Pu^{239}, 0.16% Pu^{242}, and 0.65% Pu^{241}. The U^{238} target was made of depleted U^{238} with a contamination of 0.03% U^{235}.

B. Electronics

Because of the very low counting rates in these experiments it was desirable to run at as high an incident beam current as possible. This meant that even with good coincidence-time resolution the accidental rate was large (see Sec. IIC). To make reliable corrections for accidental coincidences, it was, therefore, necessary to measure simultaneously both those events which corresponded to a real time coincidence and those to which

[8] R. Vandenbosch, J. P. Unik, and J. R. Huizenga, in *Symposium on the Physics and Chemistry of Fission, Salzburg, Austria, 1965* (International Atomic Energy Agency, Vienna, 1965).

[9] B. D. Wilkins, J. P. Unik, and J. R. Huizenga, Phys. Letters **12**, 243 (1965).

[10] A. I. Yavin and G. N. Farwell, Nucl. Phys. **12**, 1 (1959).

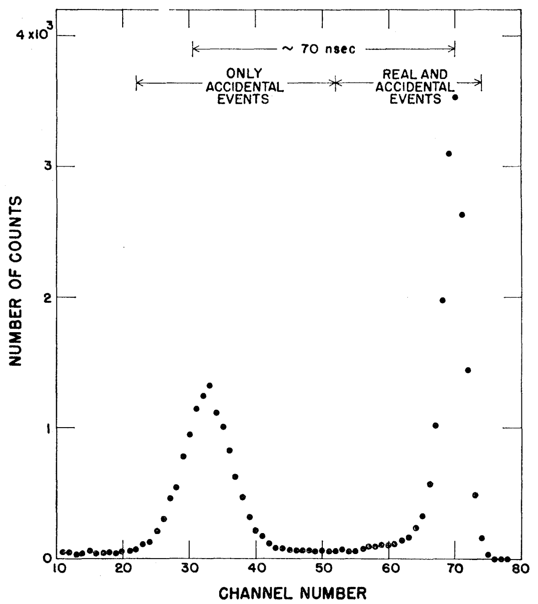

FIG. 1. Typical pulse-height spectrum from the time to pulse-height converter used in the coincidence system.

only accidental coincidences contributed because the timing of the two detectors was displaced. The coincidence information was obtained by taking a fast timing pulse from the zero crossover in a double-delay-line-clipped amplifier system[11] connected to each detector. The fast timing pulses from the alpha-particle detector were fed into one side of an overlap type time-to-height converter (THC), and the timing pulses from several fission detectors were mixed and fed into the other input of the THC. For each event the THC gave a pulse whose height was proportional to $[1-|t_D|]$, where $|t_D|$ is the absolute value of the difference between the arrival times for pulses from the fission and alpha-particle detectors. In addition, pulses were generated which could be used to identify which fission detector was involved in a given event. Variable delays in each fission channel were adjusted so that a true time coincidence corresponded to a maximum overlap between the alpha and fission timing pulses. The output from the THC and the alpha-particle pulse height were fed to two analog-to-digital converters (ADC) of a two-dimensional analyzer. Both ADC's were used with a resolution of 256 channels and, in addition, binary information was recorded to identify which fission detector was involved. The data were stored by writing the information involved in each event on magnetic tape and later sorting the relevant spectra on an IBM-7094 computer.

A typical spectrum obtained from the time-to-height converter is shown in Fig. 1. The structure in this spectrum results from the microstructure in the cyclotron beam. For 40-MeV alpha particles, beam pulses were spaced with a period of $\sim$70 nsec. The peak at channel 70 corresponds to events where both the fission fragment and the alpha particle are emitted in the same cyclotron burst. This peak is folded back on itself because of the overlap nature of the THC. The peak near channel 30 corresponds to accidental coincidences in which the fission fragment comes from the cyclotron burst immediately before or immediately after the burst in which the alpha particle originated. Thus, the spectrum of accidental events that is included in the true time coincidence peak is obtained by multiplying the spectrum of accidental coincidences by one-half. Using this timing information it is possible to make a direct correction for accidental events (see Sec. II C). The time resolution that was obtained allowed a clean separation between events occurring in adjacent cyclotron beam pulses but was not sufficient to distinguish accidental coincidences between fission fragments and alpha particles occurring within the same beam pulse. Most of the time jitter in the system was due to the slow time response of the lithium-drifted silicon detectors used for alpha-particle detection.

C. Data Analysis

As described in the previous section, the experimental data were recorded event by event on magnetic tape. The information recorded for each event consisted of digitized pulse heights for the alpha-particle energy and the output of the THC coincidence system. In addition, binary information was recorded to identify which of several fission detectors was involved in a given event. A digital computer was then used to sort separate alpha-particle pulse-height spectra for events involving each particular fission detector. For each detector two alpha-particle spectra were sorted corresponding to two pulse-height intervals in the output of the THC. These two intervals are indicated schematically in Fig. 1. The alpha-particle spectrum containing both real and accidental events, corresponds to the detection of a fission fragment and an alpha particle, both of which originated during the same cyclotron beam pulse. The interval containing only accidental events in Fig. 1 corresponds to the case where a fission fragment and an alpha particle came from adjacent beam pulses. The spectrum of real coincident events could then be obtained by subtracting one-half of the accidental events in each channel from the real plus accidental events in the corresponding channel. As a result of this analysis procedure, alpha-particle pulse-height spectra were obtained for real coincident events in each fission detector.

Because of the nature of the inelastic alpha-particle spectra, it was possible to make a direct test of the reliability of the accidental corrections described above. Since these nuclei have positive fission thresholds, it is not possible to get a real coincident event corresponding to an excitation energy significantly less than the fission

[11] F. S. Goulding and D. Landis, Natl. Acad. Sci.—Natl. Res. Council, Publ. 1184.

threshold. However, accidental events could occur for all residual excitation energies and, in particular, many accidental events were observed which corresponded to zero excitation energy or elastic scattering. If the accidental correction is made correctly, then, to within the statistical accuracies of the data, the number of elastic events in the accidental correction should equal the number of elastic events in the real plus accidental spectrum. Tests of this type indicated that the accuracy of the accidental corrections was better than 5% of the accidental rate. The accidental coincidence rates were always less than 50% and 25% of the real coincidence rates for the U^{238} and Pu^{240} measurements, respectively. This meant that the uncertainties in the final results were small compared to statistical uncertainties and to the uncertainties in normalization of detector solid angles that are described below.

One of the difficulties in obtaining the final angular correlations was to obtain the relative effective solid angles of the various detectors placed at different laboratory angles. The effective solid angle subtended by a given detector was dependent not only on its active area but also on the exact position of the beam and target relative to the center of the scattering chamber. If the beam line or the target position was displaced as little as $\frac{1}{32}$ in. from the center of the chamber, a significant change in the solid angle of a particular detector could result. The following method was used to normalize the solid angles of the various detectors. First, the laboratory angular distribution of fission fragments was measured with a fission detector placed 6 in. from the target. At a distance of 6 in., the solid angle of the detector was not significantly altered by effects due to the beam or target being slightly off the center of the chamber. Then for each coincidence run the total number of fission fragments incident on each detector was recorded. The data could then be normalized by comparing the measured fission singles to the previously measured laboratory angular distribution of fission events. This empirical normalization was checked by making detailed measurements of the exact position of the beam and target relative to the center of the chamber and measurements of the relative area of the detectors used. By comparing the normalizations obtained by these two methods and the reproducibility of various measurements, it is estimated that relative normalizations are accurate to better than 5%. The accuracy of the determination of the exact position of the beam and target also introduced an uncertainty of $\pm 1°$ in the laboratory angles of the detectors.

In order to interpret the experimental angular correlations it was necessary to transform the results to the appropriate center-of-mass system, obtain an energy calibration for the alpha-particle pulse-height spectra, and correct the results for the finite angular resolution of the detectors. The data were transformed to the rest system for the recoiling fissioning nucleus by methods described previously.[7] An energy calibration was obtained by measuring the pulse height for known alpha-particle groups from O^{16} and C^{12} at various scattering angles. Angular resolution corrections were made to the final results by graphically determining the mean angle of detection averaged over the angular distribution for each detector. This correction was done in an iterative manner and the corrected mean angles for each detector are accurate to within $\pm 2°$ relative to the symmetry angle for the angular correlation.

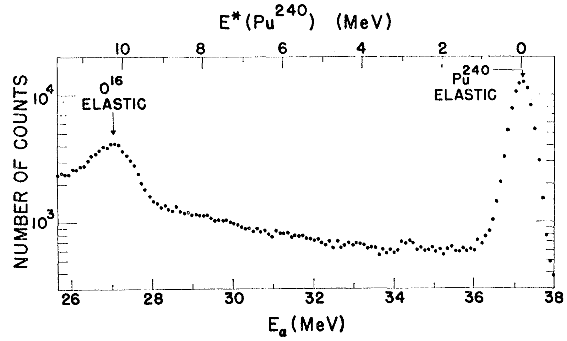

FIG. 2. Typical energy spectrum of alpha-particle singles events from the $Pu^{240}(\alpha,\alpha')$ reaction at $\theta_\alpha = 75°$.

III. EXPERIMENTAL RESULTS

A typical spectrum of inelastic alpha particles at an angle of 75° for the Pu^{240} target is shown in Fig. 2. The singles spectrum for the U^{238} target was very similar. Figure 2 illustrates the advantages of placing the alpha detector at 75°. In this case there is no C^{12} and O^{16} contamination in the spectrum in the region of excitation from 0 to 9 MeV in the residual Pu^{240} nucleus. In addition, it is possible to make measurements up to an excitation of $\sim$12 MeV because the O^{16} elastic scattering is not very intense. In fact, if the solid angle is decreased and the scattering angle optimized, it is possible to almost completely eliminate elastic scattering from O^{16} because of the deep minimum in the elastic-scattering cross section near 75°.[10] With present techniques it is not possible to make measurements for excitations greater than 12 MeV because of the intense group that occurs from elastic scattering on C^{12}.

In the coincidence measurements, angular correlations were measured by placing fission detectors at a variety of angles in the scattering plane. In addition, for the U^{238} target, measurements were made of the azimuthal angular correlation. Figure 3 shows coincident spectra observed at angles parallel and perpendicular to the kinematic recoil direction for the U^{238} and Pu^{240} targets. In these experiments the energy resolution was not sufficient to resolve individual threshold groups of the type seen in (d,pf) experiments.[7,12] The vertical lines in the Pu^{240} spectra represent the position of the two thresholds observed by Northrop,

[12] J. A. Northrop, R. H. Stokes, and K. Boyer, Phys. Rev. **115**, 1277 (1959).

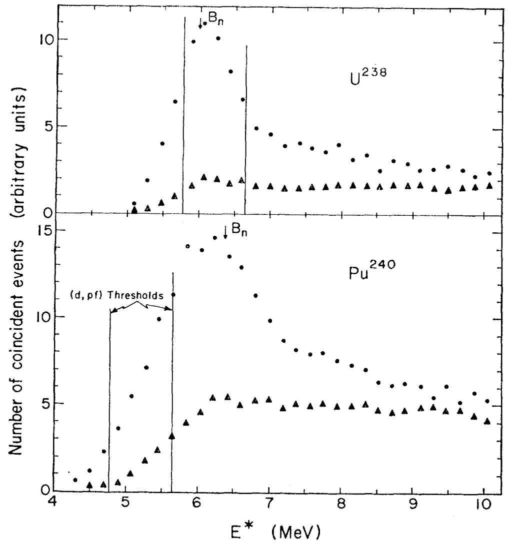

FIG. 3. Coincident energy spectra for the $(\alpha,\alpha'f)$ reaction for fission fragments observed at 0° (circles) and 90° (triangles) relative to the symmetry axis. The arrows indicate the neutron-binding energies (B_n) for the two cases. Vertical lines indicate the two thresholds determined in Ref. 12 for Pu^{240}.

Stokes, and Boyer[12] in $Pu^{239}(d,pf)$ experiments. The vertical lines for the U^{238} spectrum represent positions which are inferred for the U^{238} thresholds from a comparison of the anisotropies observed for U^{238} and Pu^{240}. The variation in the anisotropy as a function of excitation was essentially the same for the U^{238} and Pu^{240} measurements except for a shift of 1.0±0.1 MeV (see Fig. 4). This shift gives a measure of the difference in the fission thresholds for the two nuclei. In these experiments the observed anisotropies are 2 to 3 times greater than those observed in the (d,pf) experiments.[7] These anisotropies have not been corrected for the finite angular resolution of the detectors, so that near threshold the true anisotropies are somewhat greater. Because of the poor energy resolution it was not possible to resolve structure in the anisotropy as a function of excitation energy of the type observed in $Pu^{239}(d,pf)$ experiments.[7]

Azimuthal angular correlations for the $U^{238}(\alpha,\alpha'f)$ reaction were measured in a coordinate system in which the Z axis is taken along the direction of the symmetry angle (θ_s) for the in-plane angular correlation measurements. In this coordinate system measurements were made for a polar angle of 90° at azimuthal angles, ϕ, of 0°, 45°, and 90°, where $\phi=0°$ corresponds to the detector being in the reaction plane. The results in Fig. 4 show that the angular correlations are independent of the azimuthal angle to within the accuracy of the measurements.

In order to obtain reasonable statistical accuracy, the data for the full angular correlations were sorted in excitation energy intervals of 0.57 MeV, which is slightly greater than the energy resolution of the measurements. The angular correlations for the first four excitation energy intervals for the U^{238} and Pu^{240} measurements are shown in Figs. 5 and 6, respectively. These results have been transformed to the rest system for the fissioning nucleus but have not been corrected for angular resolution. Points that were taken near $\theta-\theta_R=180°$ have been reflected to show the internal consistency of the measurements and the data transformations. From these measurements it was found that the symmetry angle for the angular correlations was 4°±2° less than the calculated kinematic recoil angle. The U^{238} results are in good agreement with similar results reported by Wilkins, Unik, and Huizenga.[9]

IV. ANALYSIS AND INTERPRETATION OF RESULTS

A. Theoretical Angular Correlations

In general, if a state is formed with a total angular momentum of J and then fissions through a transition state with a value of K for the projection of the total angular momentum J on the nuclear symmetry axis, the angular distribution of the fragments can be expressed[5,6] in terms of symmetric top wave functions

$$W_{J,K}(\theta,\phi)=[(2J+1)/8\pi^2]\,|\sum_M a_M D_{MK}{}^J(\theta,\phi,\psi)|^2, \quad (1)$$

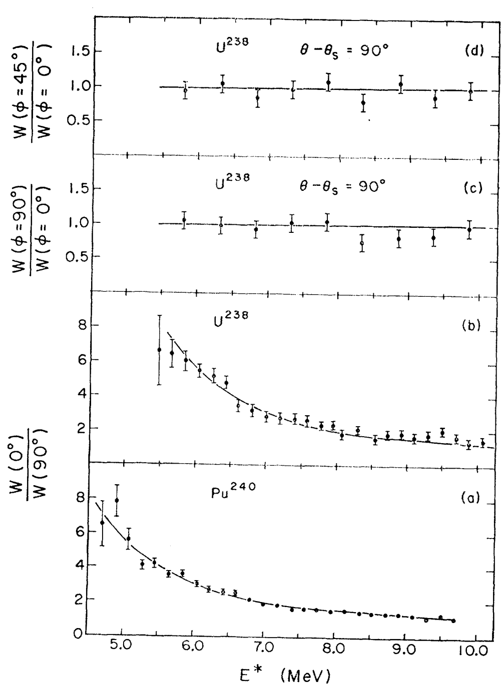

FIG. 4. Anisotropies in the fission-fragment angular correlation as a function of the excitation of the fissioning nucleus. All angles are measured relative to the symmetry direction for the angular correlation in the reaction plane. The solid curves in (a) and (b) have the same shape except for a shift of 1.0 MeV due to the difference in the fission thresholds for the two reactions. The results have not been corrected for the finite angular resolution of the detectors.

where M is the projection of J on an arbitrary Z axis and the coefficients, a_M, represent the relative amplitudes of the possible M substates. In principle, if the relative amplitudes a_M are known for all possible transition states (J,K), the total angular correlation for the $(\alpha,\alpha' f)$ reaction could be obtained by summing Eq. (1) over all possible states. In practice there is usually not enough information available to allow this general approach. However, in a special case where a Z axis can be found such that only $M=0$ substates contribute for all J values (i.e., $a_M=0$ for all $M\neq 0$), a much simplified angular correlation results[13]:

$$W_{J,K}(\theta)=|Y_J{}^K(\theta,\phi)|^2. \qquad (2)$$

In a direct reaction this special case corresponds to the plane-wave approximation with the Z axis taken in the direction of the kinematic recoil of the residual nucleus.

One consequence of the plane-wave approximation is the prediction that the azimuthal angular correlation is isotropic. The azimuthal correlations described in the previous section showed an isotropic distribution for the $U^{238}(\alpha,\alpha' f)$ reaction. These measurements provide a necessary but probably not sufficient condition for the validity of the plane-wave approximation. In order to further investigate the validity of the plane-wave approximation, DWBA calculations were performed for the $U^{238}(\alpha,\alpha' f)$ reaction and these calculations were compared to plane-wave predictions in a manner similar to that used in the analysis of (d,pf) results.[7] The details of these calculations are given in Appendix I. For inelastic alpha particles observed at 75°, except for a small difference in the symmetry angle, the DWBA

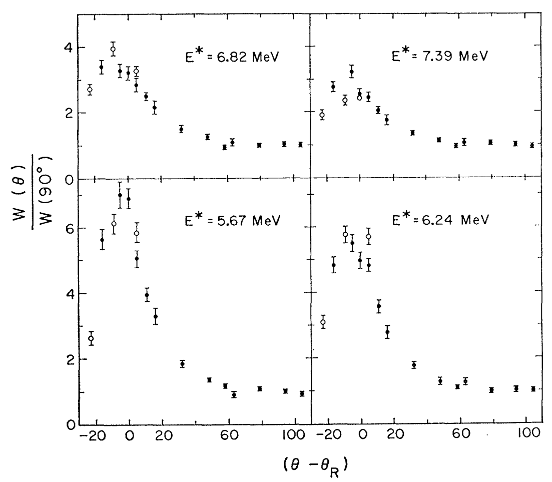

FIG. 5. Angular correlations of fission fragments from the $U^{238}(\alpha,\alpha' f)$ reaction. The data have been sorted in excitation energy intervals of 0.57-MeV width. Open circles indicate points which have been reflected by 180°. The angular scale is relative to the kinematic recoil angle θ_R. The results have not been corrected for finite angular resolution of the fission detectors.

[13] For a discussion of the properties of the $D_{M,K}{}^J$ functions, see M. A. Preston, *Physics of the Nucleus* (Addison-Wesley Publishing Company, Inc., Reading, Massachusetts, 1962), Appendix A-2.

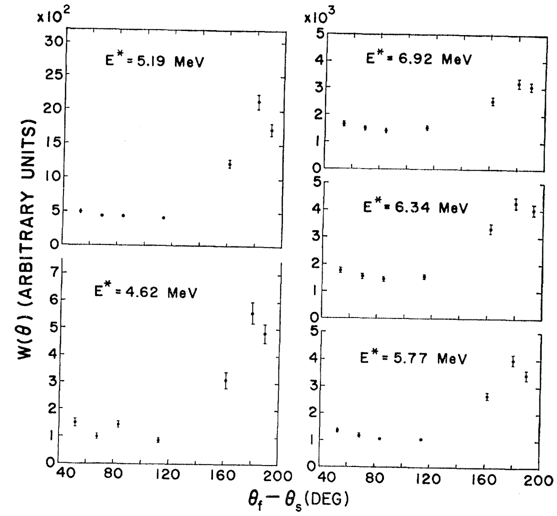

FIG. 6. Angular correlations of fission fragments from the $Pu^{240}(\alpha,\alpha' f)$ reaction. The data have been sorted in excitation energy intervals of 0.57-MeV width. The angular scale is relative to the kinematic recoil angle θ_R. The results have not been corrected for the finite angular resolution of the fission detectors.

calculations yielded the same angular correlations as plane-wave calculations for a given J,K. These calculations indicate that if the Z axis is taken along the symmetry axis of the angular correlation it is a good approximation to assume that only $M=0$ substates are populated.

Assuming that only $M=0$ substates are populated, the angular correlation in the reaction plane for a given J,K from Eq. (2) is

$$W_{J,K}(\theta)=\frac{2J+1}{2}\frac{(J-K)!}{(J+K)!}|P_J{}^K(\cos\theta)|^2\,,$$

where the quantities $P_J{}^K(\cos\theta)$ are associated Legendre functions defined as

$$P_J{}^K(\cos\theta)=\sin^K\theta\left(\frac{\partial^K}{(\partial\cos\theta)^K}P_J(\cos\theta)\right).$$

The total observed angular correlation can be written as

$$W(\theta)=\sum_K A_K\sum_J \sigma(J)N(J)W_{J,K}(\theta). \qquad (3)$$

In Eq. (3) it is assumed that the transition-state spectrum is made up of rotational bands built on vibrational states of a specific K projection. Then A_K represents the relative probability of fissioning through all of the available transition states of a given K. This form assumes that the relative probability of fission through a given vibrational band is independent of the total angular momentum J for the transition state involved except that only natural parity states are allowed. The characteristics of the initial direct reaction are described by $\sigma(J)$ and $N(J)$. The probability of exciting a level with a specific value of J is given by $\sigma(J)$. $N(J)$ represents an average number of levels that are available in the residual nucleus formed by the (α,α') direct reaction.

For the (α,α') reactions, the projectile and target spins are zero so that $\sigma(J)$ is given by the cross sections for various angular-momentum transfers. These cross sections are obtained from the DWBA calculations described in Appendix I. Furthermore, the (α,α') reaction can excite only natural parity states so that the sum on J in (3) is taken over only even J values for positive-parity transition states and over only odd J values for negative-parity transition states. The results of the DWBA calculations indicated that for $\theta_{\alpha'}=75°$ and $Q=-6$ MeV $\sigma(J)$ was approximately proportional to $2J+1$ for $0\leq J\leq 10$ and decreased rapidly for $J>10$. In the fits to the experimental data $\sigma(J)$ was taken proportional to $(2J+1)$ and the summations were taken only up to $J=10$.

At present there is little theoretical or experimental evidence available on the form of $N(J)$. Fits were made to the experimental data assuming two empirical forms for $N(J)$.

$$N_1(J)=(2J+1)\exp[-J(J+1)/C^2],\qquad N_2(J)=\exp[-J(J+1)/C^2]. \tag{4}$$

If C^2 is interpreted as the spin cutoff factor in the total level density, then $N_1(J)$ assumes that the distribution of levels as a function of angular momentum transferred in the (α,α') reaction has the same form as the over-all distribution of levels in the residual nucleus. In fitting the experimental results the form $N_2(J)$ was used mainly in an attempt to determine how sensitive the conclusions were to the form assumed for $N(J)$.

B. Method of Data Analysis

In the analysis of the experimental angular correlations a least-squares fitting code was developed using the form given in Eq. (3). In these fits the relative probabilities of fission through transition states with different K values, A_K, were treated as adjustable parameters, and the vibrational bands which could be used in the fitting were $K=0^+, 0^-, 1^-, 2^+, 3^-, 4^+$. These represent the vibrational bands which are expected to occur at relatively low excitation energy in the transition-state spectrum.[2,3,14,15] In addition, the cutoff factor C in the density of levels $N(J)$ was treated as an adjustable parameter.

The procedure used in fitting the experimental results was to perform several least-squares fits to each angular correlation assuming that various possible combinations of the above vibrational bands were accessible for fission at a particular excitation energy. Then the following criteria were used to decide which combinations gave acceptable fits:

(1) Weighted variance. For each fit a weighted variance, $\chi^2/[(\text{No. of points})-(\text{No. of parameters})]$, was calculated. In judging the best fit for each case it was required that this weighted variance be a minimum.[16]

(2) C values independent of E^*. Since the values of C are meant to describe a distribution of levels excited in the direct reaction, it is not expected that the character of this distribution should change appreciably over the rather limited excitation energy range (~5–8 MeV) being considered.

(3). Reasonableness of A_K values. Several criteria can be placed on the reasonableness of the A_K values obtained from a given fit. If a channel description of the fission process is assumed, then at a particular excitation energy the nucleus can proceed to fission with roughly equal probability through any transition state that lies below the excitation energy available. This means that the vibrational bands allowed in fitting a given excitation energy group must include all of the bands that were needed to fit the results at excitation energies below this group. Furthermore, the ratio of the values obtained for A_K gives a measure of the relative number of bands that are present in the transition-state spectrum, and it is expected that at these low excitation energies there should be relatively few bands available with a given value of K.

Before describing the detailed fits it is interesting to discuss a few general properties of the functional form used to fit the angular correlations. Because the (α,α') reaction excites natural parity levels for a given band, only the even or the odd angular-momentum states are excited for bands of positive and negative parity, respectively. The functions $W_{J,K}(\theta)$ have the property that they give a local maximum at $\theta=90°$ if $J-K$ is even and they are zero at $\theta=90°$ if $J-K$ is odd. This means that in principle it is possible to distinguish between positive- and negative-parity bands of the same K by carefully measuring the angular correlation near 90°. In the present results the points available near 90° are not spaced closely enough to distinguish the parity of the band so that in all the fits there is an ambiguity as to the parity of the bands which are present. As a result, the parity of a band that is observed in the angular correlation can be reliably assigned only in the case where it is the lowest lying band with a given K

[14] It is possible that other vibrational bands such as $K=1^+$, 2^-, etc., could occur in the transition-state spectrum but these would have to come from multiphonon excitations and would, therefore, lie somewhat higher in excitation energy. For a more complete discussion see Ref. 15.

[15] J. J. Griffin, in *Symposium on the Physics and Chemistry of Fission, Salzburg, Austria, 1965* (International Atomic Energy Agency, Vienna, 1965).

[16] In a good fit to a series of points with known uncertainties the weighted variance should have a value near one. In this work the fits obtained to the angular correlations give weighted variances which are always significantly greater than one. This indicates that either the uncertainties assigned to the points are too small or that the form of the fitted function is not quite appropriate for the data. The fitted function could easily be slightly incorrect because of the arbitrary choice, which had to be made for the form of $N(J)$.

(e.g., the first $K=0$ band should have positive parity, the first $K=1$, negative parity, etc.).

It was found that satisfactory fits to the experimental data could be obtained using either of the two forms $N_1(J)$ or $N_2(J)$ [see Eqs. (4)] for the level density. The only essential difference in the parameters obtained from these fits was that different values for C resulted from fitting with the two forms.

C. Analysis of Experimental Results

The experimental angular correlations were determined for the U^{238} and Pu^{240} measurements for excitation energy intervals which were 0.57-MeV wide, and the intervals were centered at the same energies relative to the fission threshold for the two sets of measurements.

The first step in the analysis of the angular correlations was to calculate the best fits for each energy interval using C and various combinations of the A_K coefficients as adjustable parameters. The coefficients resulting from best fits to the U^{238} data using $N_2(J)$ are given in Table I. The first two criteria above are satisfied for the following series of fits: (1) $E^*=5.67$ and 6.24 MeV—contributions from vibrational bands with both $K=0$ and $K=1$; (2) $E^*=6.82$, 7.39, and 7.97 MeV—reasonable fits can be obtained assuming contributions from either $K=0$, 1, 2, and 4 bands or $K=0$, 1, and 3 bands. In the fitting procedure the bands are assumed to have parities $K=0^+$, 1^-, 2^+, 3^-, 4^+. However, as is described above, the fits would not be measurably changed if the opposite parity were assumed for each band. For the fits listed above, values are obtained for C in the range $C=7.6\pm0.4$, which is within the accuracy to which C can be determined. If the form $N_1(J)$ is used, a value $C=5.8\pm0.3$ is obtained from the least squares fits.

If the forms $N_1(J)$ and $N_2(J)$ are describing the spin cutoff in the nuclear level density, then the parameters C obtained from the fits should be related to the spin cutoff parameter $2\sigma^2$ by $C^2=2\sigma^2$. Calculations of σ have been performed on the basis of a superconductor model of the nucleus for $A=115$ and $A=196$ by Vonach, Vandenbosch, and Huizenga.[17] Extrapolating these results to U^{238} for an excitation of 6–8 MeV gives a qualitative prediction of $2\sigma^2\sim50$–70. The values of C^2 from the fits using $N_1(J)$ and $N_2(J)$ are 34 ± 4 and 58 ± 6, respectively.

TABLE I. Coefficients obtained from least-squares fits to $U^{238}(\alpha,\alpha' f)$ angular correlations as described in the text. A dash indicates that a particular parameter was held fixed at a value of zero for that fit. Fits were obtained using the level density for $N_2(J)$ described in the text.

E^* (MeV)	A_{0^+}	A_{1^-}	A_{2^+}	A_{4^+}	A_{3^-}	C	Weighted variance
5.67	71±11	···	···	···	···	5.0±0.4	22
	20±4	22±3	···	···	···	7.6±0.6	7.0
	20±2	23±5	0[a]	···	···	7.5±0.6	7.6
6.24	490±70	···	···	···	···	2.4±0.2	27
	21±4	37±4	···	···	···	8.0±0.6	7.2
	23±4	28±6	7±4	···	···	8.3±0.7	7.3
6.82	20±7	57±7	···	···	···	5.6±0.4	9.0
	21±3	11±5	21±3	···	···	7.3±0.5	5.7
	15±4	17±6	9±6	7±4	···	8.0±0.8	5.9
	12±3	22±4	b	b	13±2	8.2±0.8	5.6
7.39	37±13	86±10	···	···	···	4.1±0.4	8.9
	21±3	10±7	32±4	···	···	6.0±0.8	5.5
	8±3	20±5	3±5	15±3	···	7.5±0.6	4.8
	10±2	20±4	b	b	19±2	7.3±0.6	4.5
7.97	17±3	10±9	39±5	···	···	5.9±0.5	7.8
	5±3	17±6	7±5	16±4	···	7.6±0.8	7.1
	5±3	21±7	b	b	22±6	7.3±0.9	6.7

[a] If $K=2^+$ band is allowed, the fit is improved only if A_{2^+} is allowed a negative value.
[b] If $K=2^+$ and $K=4^+$ bands are allowed in these fits, then the fit is improved only with a negative value for A_{3^-}.

TABLE II. Coefficients obtained from least-squares fits to U^{238}-$(\alpha,\alpha' f)$ angular correlations as described in the text. A dash indicates that a particular parameter was held fixed at a value of zero for that fit. Fits were obtained using the level density for $N_2(J)$ described in the text. In these fits the parameter C was held fixed at a value of 7.6.

E^* (MeV)	A_{0^+}	A_{1^-}	A_{2^+}	A_{4^+}	A_{3^-}	Weighted variance
5.67	36±5	···	···	···	···	24
	20±2	22±2	···	···	···	6.5
	20±2	23±4	0±3[a]	···	···	7.1
6.24	50±4	···	···	···	···	33
	24±2	39±3	···	···	···	6.8
	27±3	31±6	7±4	···	···	6.9
6.82	8±3	36±3	···	···	···	10.7
	20±3	10±4	20±5	···	···	5.4
	16±3	17±6	11±6	6±4	···	5.5
	13±2	25±3	b	b	14±2	5.3
7.39	2±3	32±3	···	···	···	16
	15±2	4±4	23±3	···	···	6.4
	8±2	20±4	3±5	15±3	···	4.4
	9±2	20±2	b	b	18±2	4.2
7.97	13±2	0.5±4	29±3	···	···	9.1
	6±3	18±5	7±5	16±3	···	6.5
	5±2	20±2	b	b	22±2	6.2

[a] If $K=2^+$ band is allowed, the fit is improved only if A_{2^+} is allowed an unphysical value.
[b] If $K=2^+$ and $K=4^+$ bands are allowed in these fits, then the fit is improved only with a negative value for A_{3^-}.

TABLE III. Coefficients obtained from the least-squares fits to $Pu^{240}(\alpha,\alpha' f)$ angular correlations as described in the text. A dash indicates that a particular parameter was held fixed at a value of zero for that fit. Fits were obtained using the level density for $N_2(J)$ described in the text. In these fits the parameter C was held fixed at a value of 7.6.

E^* (MeV)	A_{0^+}	A_{1^-}	A_{2^+}	A_{4^+}	Weighted variance
4.62	9±7	···	···	···	16
	4±0.5	7.1±1.0	···	···	3.1
	5±1	5±3	2±2	···	3.7
5.19	37±4	···	···	···	39
	15±2	29±2	···	···	2.8
	19±2	17±4	9±3	···	2.1
5.77	18±8	85±9	···	···	11
	41±9	30±17	38±10	···	7.6
	41±14	31±25	37±25	0.5±12	10.1
6.34	14±8	120±11	···	···	20
	36±8	38±18	68±14	···	10
	31±10	56±28	40±34	18±18	13
6.92	8±9	98±14	···	···	46
	34±7	53±18	90±15	···	18
	22±8	48±25	23±30	44±18	16

[17] H. K. Vonach, R. Vandenbosch, and J. R. Huizenga, Nucl. Phys. **60**, 70 (1965).

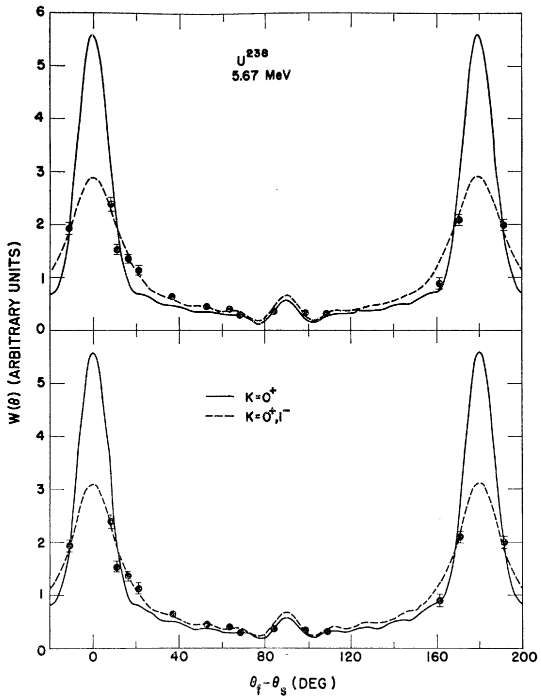

FIG. 7. The angular correlations of fission fragments from the $U^{238}(\alpha,\alpha' f)$ reaction for the excitation energy interval centered at 5.67 MeV. Experimental results have been corrected for the finite angular resolution of the detectors. The curves represent least-squares fits to the experimental data as described in the text. The curves in the top and bottom of the figure are results for fits assuming a density of levels given by $N_1(J)$ and $N_2(J)$, respectively (see text).

To examine the A_K coefficients, the next step in the analysis was to perform fits to both the U^{238} and Pu^{240} results holding the value of C fixed at 7.6 with the level density form $N_2(J)$ and using the A_K's as adjustable parameters. These results are shown in Tables II and III and the fitted curves are shown in Figs. 7–12. From Tables II and III it is seen that the fits listed above also give reasonable ratios for the A_K coefficients and the results are basically the same for the U^{238} and Pu^{240} angular correlations.

The values which are obtained for the A_K's are, of course, dependent on the detailed model for the (α,α') process which is described in Sec. IVA. In particular, if the density of levels excited by the (α,α') reaction is very different in form from that postulated in Eqs. (4), then it is possible that the true A_K coefficients could be qualitatively different from those obtained from fits with the present model.

It is interesting to compare the excitation energy intervals for these results with the positions of the steps observed in the $Pu^{239}(d,pf)$ angular correlation measurements.[7] The first step in the (d,pf) results comes approximately at the boundary between the second and third intervals (i.e., between the 6.24- and 6.82-MeV groups for U^{238}). The second (d,pf) step comes approximately in the middle of the fourth interval ($E^*=7.39$ MeV for U^{238}). The (d,pf) step corresponding to the onset of two quasiparticle excitations comes approximately 0.2 MeV above the boundary for the fifth interval which with the experimental energy resolution would allow a small amount of fission through two quasiparticle states in the fifth energy interval.

Tentative assignments of energy positions and K values of the vibrational bands present near threshold can be obtained by using the (d,pf) results[7] to establish the positions of the onsets of various bands and the $(\alpha,\alpha' f)$ results for the identification of the K values. Table IV gives the ratios of the A_K coefficients, determined in the least-squares fits, to the coefficient for the $K=0^+$ ground-state band. It is interesting to consider the results in Table IV for the excitation energy intervals corresponding to the three distinct groups observed in the (d,p) measurements. In the following discussion parity assignments can be tentatively made for some of the bands on the assumption that the first bands of a given K will have the expected parities of $K=0^+$, 1^-, 2^+, 3^-, 4^+. If a second band of a given K appears in the spectrum, it is not possible to determine its parity from the present experimental results.

The first interval corresponds to excitations from the fission threshold to $\sim$0.7 MeV above threshold and should be equivalent to the first two groups (5.67 and

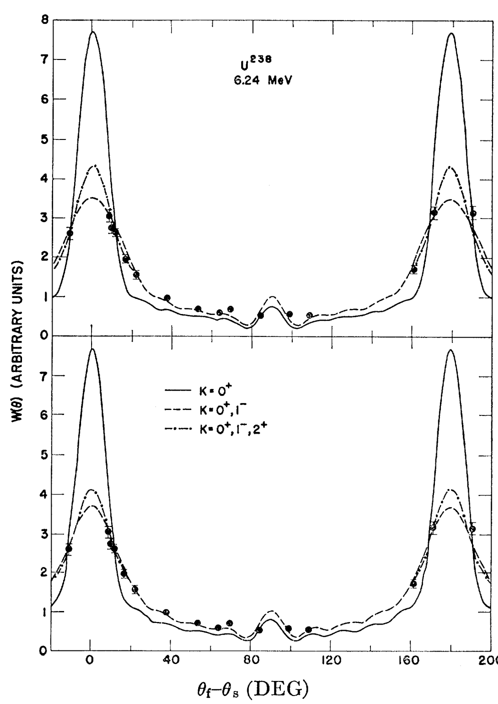

FIG. 8. The angular correlations of fission fragments from the $U^{238}(\alpha,\alpha' f)$ reaction for the excitation energy interval centered at 6.24 MeV. Experimental results have been corrected for the finite angular resolution of the detectors. The curves represent least-squares fits to the experimental data as described in the text. The curves in the top and bottom of the figure are results for fits assuming a density of levels given by $N_1(J)$ and $N_2(J)$, respectively (see text).

6.24 MeV) in the $U^{238}(\alpha,\alpha' f)$ measurements. Table IV shows that these groups can be characterized by equal contributions from $K=0$ and $K=1$ bands. In this region a reasonable interpretation is that one $K=0^+$ band and one $K=1^-$ band are present.

The second interval in the (d,pf) results corresponds to excitations from 0.7 to 1.6 MeV above threshold and would be characterized best by the third group (6.82 MeV) in the $U^{238}(\alpha,\alpha' f)$ measurements. Table IV indicates that there are two equivalent fits to the data in this energy interval: (1) There are equal contributions to fission from $K=0$, 1, and 2 bands, or (2) there are contributions from $K=0$, 1, and 3 bands with intensity ratios of approximately 1:2:1. These results suggest that at an excitation energy of $\sim$0.7 MeV the transition-state spectrum contains either (1) a $K=2^+$ band or (2) a combination of a $K=3^-$ band and an additional $K=1$ band.

The third interval corresponds to excitations above threshold which are between $\sim$1.6 MeV and the onset of two quasiparticle excitations at $\sim$2.6 MeV. This interval includes the upper half of the fourth group (7.39 MeV) and the fifth group (7.97 MeV) in the $U^{238}(\alpha,\alpha' f)$ results. For these groups the A_K coefficients are becoming quite uncertain and again there are two equivalent fits to the experimental results: (1) There are contributions from $K=0$, 1, 2, and 4 bands with intensity ratios of the order 1:2:1:2; or (2) there are contributions from $K=0$, 1, and 3 with intensity ratios of the order of 1:2:2. The simplest interpretation of these results would be that, at approximately 1.6 MeV, the transition state spectrum contains either (1) two $K=4$ bands and an additional $K=1$ band or (2) an additional $K=3$ band. The results in Table IV for the 7.97-MeV U^{238} group seem to indicate a more complex structure but this is very uncertain because of the possible inclusion in the spectrum of two quasiparticle states due to both the poor energy resolution of the present experiment and the possibility that the pairing gap could be less for the U^{238} saddle shape than for Pu^{240}.

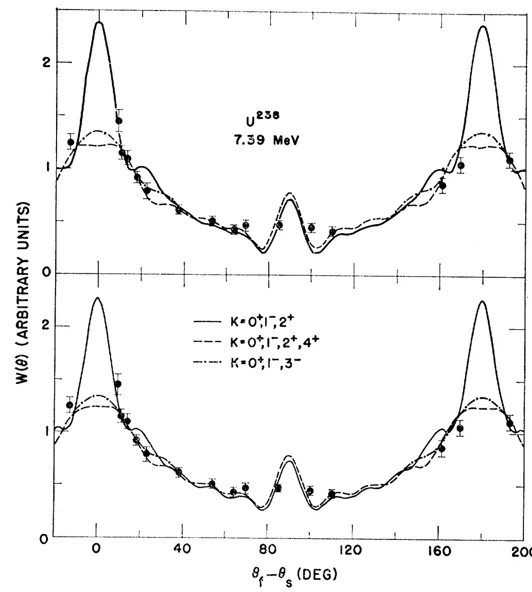

FIG. 10. The angular correlation of fission fragments from the $U^{238}(\alpha,\alpha' f)$ reaction for the excitation energy interval centered at 7.39 MeV. Experimental results have been corrected for the finite angular resolution of the detectors. The curves represent least-squares fits to the experimental data as described in the text. The curves in the top and bottom of the figure are results for fits assuming a density of levels given by $N_1(J)$ and $N_2(J)$, respectively (see text).

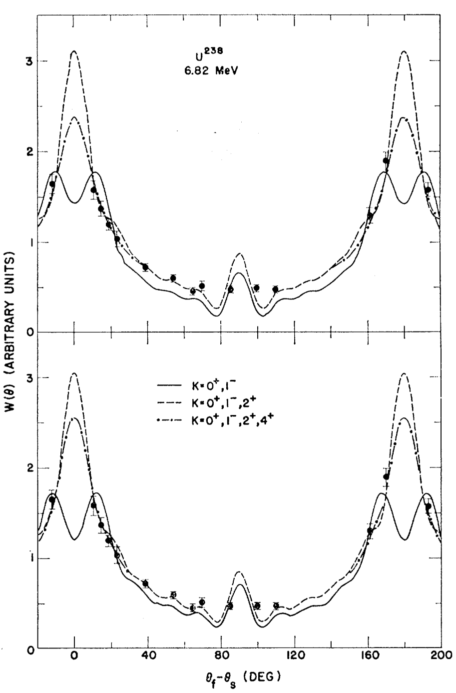

FIG. 9. The angular correlations of fission fragments from the $U^{238}(\alpha,\alpha' f)$ reaction for the excitation energy interval centered at 6.82 MeV. Experimental results have been corrected for the finite angular resolution of the detectors. The curves represent least-squares fits to the experimental data as described in the text. The curves in the top and bottom of the figure are results for fits assuming a density of levels given by $N_1(J)$ and $N_2(J)$, respectively (see text).

V. DISCUSSION

The results from the previous section indicated that the present $(\alpha,\alpha' f)$ results and positions for the structure in (d,pf) anisotropy measurements[7] could be used to generate two possible energy level diagrams for the transition-state spectrum of Pu^{240} and U^{238} which are consistent with the $(\alpha,\alpha' f)$ angular correlations. A schematic representation of these energy level diagrams is shown in Fig. 13.

A check on the reliability of these conclusions is to reanalyze the previous (d,pf) anisotropies[7] assuming the transition-state spectra shown in Fig. 13. This comparison was accomplished by theoretically calculating the coefficient $\bar{g}_2$ in the expansion $W(\theta)\propto 1+\bar{g}_2 P_2(\cos\theta)$

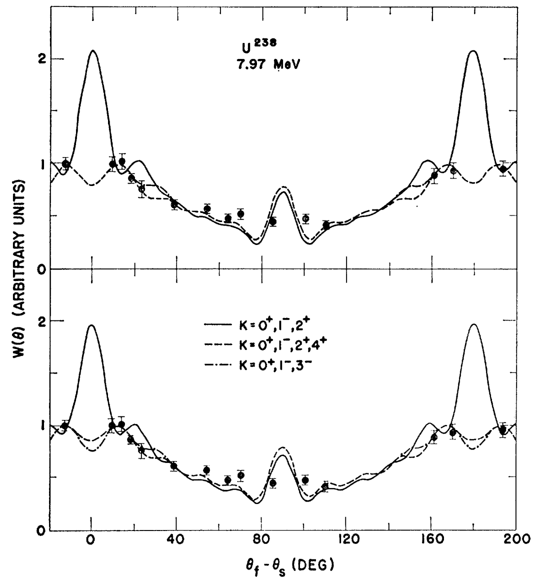

FIG. 11. The angular correlations of fission fragments from the $U^{238}(\alpha,\alpha'f)$ reaction for the excitation energy interval centered at 7.97 MeV. Experimental results have been corrected for the finite angular resolution of the detectors. The curves represent least-squares fits to the experimental data as described in the text. The curves in the top and bottom of the figure are results for fits assuming a density of levels given by $N_1(J)$ and $N_2(J)$, respectively (see text).

assuming that various vibrational bands are available. Details of these calculations are given in Appendix II. A comparison of these calculations with the experimental (d,pf) results[7] is shown in Fig. 14. Figure 14 shows that the two possible level schemes which were obtained from the $(\alpha,\alpha'f)$ results are also consistent with the (d,pf) measurements.

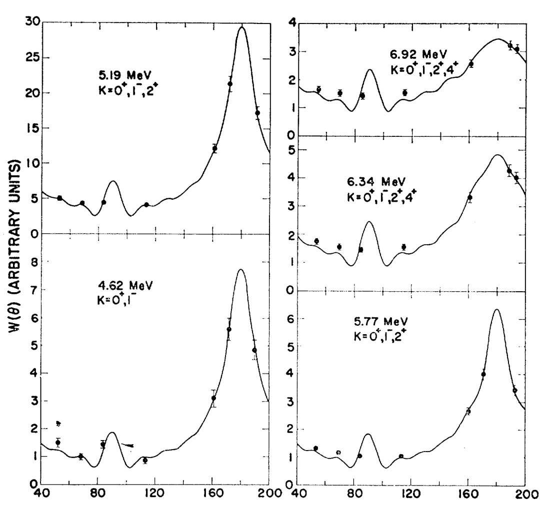

FIG. 12. The angular correlations of fission fragments from the $Pu^{240}(\alpha,\alpha'f)$ reaction. Experimental results have been corrected from the finite angular resolution of the fission detectors. Solid curves show least-squares fits to the data using the form $N_2(J)$ for the density of excited levels.

It is possible to obtain qualitative estimates of the positions of various vibrational bands in the transition state spectrum of an even-even nucleus from calculations of the frequencies of various vibrational modes using a liquid-drop model of the nucleus. A rough estimate of this type has been made by Wheeler.[3] These estimates predict a $K=0^-$ band in the region $0.1<E^*-E_f<0.6$ MeV; a $K=1^-$ band in the region $0.5<E^*-E_f<1.5$ MeV; and a $K=2^+$ band at $E^*-E_f\sim 1$ MeV. The level spectrum in Fig. 13I is in approximate agreement with these estimates except for the apparent absence of a $K=0^-$ band.

As discussed above, the fits to the $(\alpha,\alpha'f)$ results are not very sensitive to the parity of the vibrational bands which are contributing. However, if for the lowest excitation energy interval there were contributions from a $K=0^-$ band in addition to $K=0^+$ and $K=1^-$ bands, then one would expect[18] the ratio $A(K=1)/A(K=0)$ to be $\sim\frac{1}{3}$ instead of the value of ~ 1 which is observed. Similarly, the interpretation of the A_K coefficients for the higher excitation energy intervals becomes complicated. In order to explain the observed results, it would be necessary to postulate several bands with each possible K value. In the (d,pf) results the addition of a $K=0^-$ band gives a poorer fit to the experimental data unless a complicated band structure is assumed. If the transition-state spectrum is much more complicated than that shown in Fig. 12, it seems unlikely that only two distinct steps would be observed in the (d,pf) anisotropies. Thus, the data from both these experiments are most easily interpreted if it is assumed that there is no contribution from a low-lying $K=0^-$ vibrational band. In principle, it should be possible to make a definite determination of the parities of the vibrational bands contributing at a given excitation energy from a careful investigation of the $(\alpha,\alpha'f)$ angular correlation near 90°.

Recently, it has been postulated by Griffin[15] that there should exist in the Pu^{240} transition-state spectrum a vibrational band with $K=1^+$ at an excitation energy of $E^*-E_f\leq 1.6$ MeV. This band is needed to explain the resonance neutron fission of Pu^{239} through 1^+ levels. The two possibilities for the transition-state spectrum (Fig. 13) both indicate a $K=1$ band which is in addition to the lowest $K=1$ band. The second band could be a $K=1^+$ band, and again it should be possible to determine the parity by careful measurements of the $(\alpha,\alpha'f)$ angular correlation near 90°.

In the analyses for both experiments the results are sensitive to the assumptions which are made concerning the initial direct reaction. For both cases one major assumption is that for a specific angular momentum

[18] If the fission near threshold involves $K=0^+, 0^-$, and 1^- transition state bands, then one would expect the cross section for exciting negative parity states to split between $K=0^-$ and 1^- bands, and the cross section for exciting positive parity bands to lead to fission entirely through the $K=0^+$ band. If this were true, then the ratio $A(1)/A(0)$ should be $\sim\frac{1}{3}$.

TABLE IV. Ratios of the coefficients obtained in the least-squares fits listed in Tables II and III.

E^* (MeV)	Target	$A(1^-)/A(0^+)$	$A(2^+)/A(0^+)$	$A(4^+)/A(0^+)$	$A(3^-)/A(0^+)$
5.67	U	1.1±0.2	0.0	...	...
4.62	Pu	0.9±0.3	0.3±0.3	...	...
6.24	U	1.1±0.2	0.2±0.1	...	...
5.19	Pu	0.9±0.3	0.5±0.2	...	...
6.82	U	1.1±0.3	0.7±0.3	0.4±0.3	...
6.82	U	1.9±0.2	...	...	1.0±0.2
5.77	Pu	0.8±0.6	0.9±0.6	0.0±0.3	...
7.39	U	2.5±0.5	0.3±0.8	1.9±0.4	...
7.39	U	2.2±0.3	...	...	2.1±0.3
6.34	Pu	1.8±0.8	1.3±1.0	0.6±0.6	...
7.97	U	3.2±1.0	1.2±1.0	2.9±0.8	...
7.97	U	4.0±0.5	...	...	4.4±0.5
6.92	Pu	2.2±1.0	1.0±1.0	2.0±1.0	...

transfer the angular correlations can be calculated from a plane-wave theory. For both reactions more exact DWBA calculations have indicated that this approximation is appropriate for the specific conditions of the experiments. In addition for the $(\alpha,\alpha' f)$ reaction, measurements indicated that there is no azimuthal dependence for the angular correlation which is a consequence of the plane-wave approximation.

The other major approximation which is involved in the analysis of both the $(\alpha,\alpha' f)$ and the (d,pf) data is the form of the distribution of angular-momentum states excited in the direct reaction $[N(J)]$. In the $(\alpha,\alpha' f)$ case it was assumed that the distribution of levels was of approximately the same statistical form as the distribution of levels which exist in the excited Pu^{240} or U^{238} nuclei. At present, because of the lack of experimental information on the relative excitation of levels in (α,α') reactions at the high excitations, the statistical assumption which was used seems most reasonable. However, it is possible that specific effects such as a strong excitation of 1^- levels due to the tail of the giant dipole resonance could perturb the $N(J)$ distribution considerably. Such effects could then lead to a quite different interpretation of the $(\alpha,\alpha' f)$ angular correlations. The major evidence that the analysis of the $(\alpha,\alpha' f)$ results is approximately correct is the agreement between these results and the analysis of the (d,pf) measurements.[7] In the analysis of the (d,pf) data it was assumed that $N(J)=\text{constant}$. This assumption was based on the properties of the single particle levels available for the stripping process[7] and is not

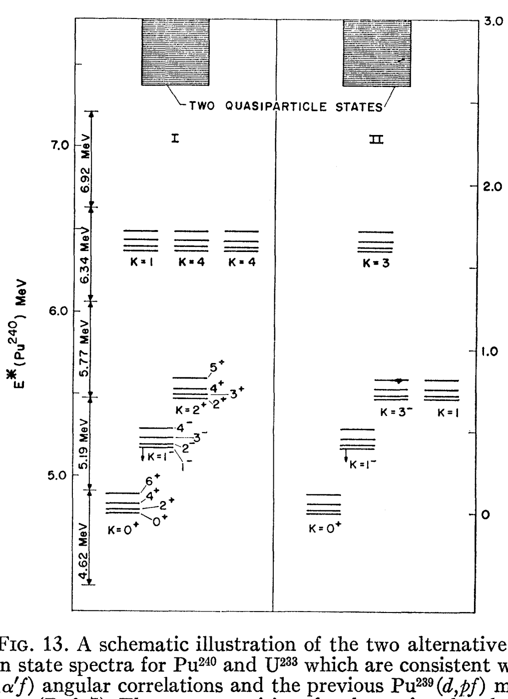

FIG. 13. A schematic illustration of the two alternative transition state spectra for Pu^{240} and U^{233} which are consistent with the $(\alpha,\alpha' f)$ angular correlations and the previous $Pu^{239}(d,pf)$ measurements (Ref. 7). The energy positions for the various bands are determined approximately from the $Pu^{239}(d,pf)$ measurements and the values of K for each band from the $(\alpha,\alpha' f)$ angular correlations. Only the first few levels of each band are indicated and the rotational spacings are not drawn to scale. The energy intervals corresponding to the angular correlations shown in Fig. 12 are indicated by arrows.

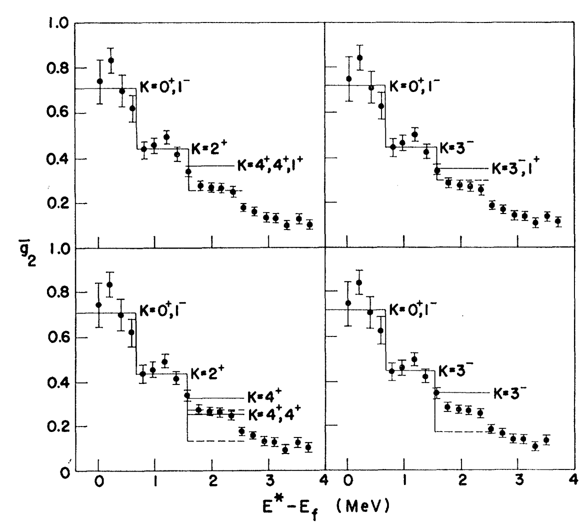

FIG. 14. Results for the coefficient $\bar{g}_2$ in the angular correlation, $W(\theta)\propto 1+\bar{g}_2P_2(\cos\theta)$, for the $Pu^{239}(d,pf)$ reaction. The experimental results are taken from Ref. 2. Solid lines indicate results of calculations assuming $\Gamma_f(J)\sim\Gamma_T(J)$. Dashed lines indicate results of calculations assuming $\Gamma_f(J)\ll\Gamma_T(J)$.

subject to the same uncertainties as the statistical assumption used in the $(\alpha,\alpha' f)$ analysis. It seems unlikely that the quite different assumptions used to characterize the (α,α') and (d,p) reactions could both be qualitatively in error in such a way as to give agreement for the transition state spectrum implied from the two experiments.

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APPENDIX I. DWBA CALCULATIONS

In the analysis of the $(\alpha,\alpha' f)$ results it was assumed that for a given angular momentum transfer the angular correlation of the fragments could be described by a plane-wave theory or, equivalently, if a coordinate system is chosen with the Z axis in the direction of the symmetry angle for the angular correlation, only $M=0$ states are excited. In order to check the validity of this approximation, DWBA calculations were performed for the (α,α') reaction and the results compared to plane-wave predictions. The DWBA calculations were performed using the T–SALLY code[19] with optical-model parameters obtained from Igo.[20] These calculations were performed using two different choices for the interaction form factor: (1) a first derivative of the Saxon well and (2) the second derivative of the Saxon well.

From the results of these calculations angular correlations for the emitted fragments can be calculated in a manner analogous to that previously described[7] for the (d,pf) reaction. These calculations were performed as follows: For a specific angular-momentum transfer l to a state with excitation energy Q corresponding to an inelastic alpha particle emitted at an angle θ_α, the T–SALLY code was used to obtain values of β_l^m. Then the angular correlation of the fission fragments in the reaction plane for the condition $K=0$ is given by

$$W_l(\theta_f)\propto \sum_{LM}(2L+1)^{-1/2}g(Ll0)\tilde{\rho}_{Ll}{}^M(\mathbf{k}_p)Y_L{}^M(\theta_f)\,,\quad (\text{A1})$$

where

$$\tilde{\rho}_{Ll}{}^M=\frac{\sum_{mm'}(LlMm|Lllm')\beta_l{}^m(\mathbf{k}_p)\beta_l{}^{m'}(\mathbf{k}_p)^*}{(Ll00|Ll l0)\sum_m|\beta_l{}^m(\mathbf{k}_p)|^2}\,,\quad (\text{A2})$$

$$g(Ll0)=(2L+1)|(Ll00|Ll l0)|^2.\quad (\text{A3})$$

In these calculations θ_f is the angle of emission of a fission fragment referred to the beam axis. In the plane-wave approximation the angular correlation of the fragments is obtained by using

$$\tilde{\rho}_{Ll}{}^M=(4\pi/(2L+1))^{1/2}Y_L{}^M(\theta_R)\,,\quad (\text{A4})$$

where θ_R is the kinematic recoil angle for the fissioning nucleus.

Plane-wave and DWBA angular correlations were calculated for various alpha-particle angles θ_α and $l=1$, 2, 3 for $K=0$. Results of these calculations for $\theta_\alpha=30°$, 45°, 60°, and 75° are shown in Fig. 15. The DWBA results in Fig. 15 are for a form factor proportional to the first derivative of a Saxon well. The results obtained with the second-derivative form factor were very similar and for $\theta_\alpha>45°$ were essentially identical. Figure 15 shows that as θ_α is increased the DWBA correlations more closely approximate the predictions of a plane-wave theory. At $\theta_\alpha=75°$, the condition for this experiment, the plane-wave and DWBA predictions are essentially identical for $l\leq 3$ except for a difference of $\sim 2°$ in the symmetry angle predicted in the two cases. This 2° deviation is the same for all l values

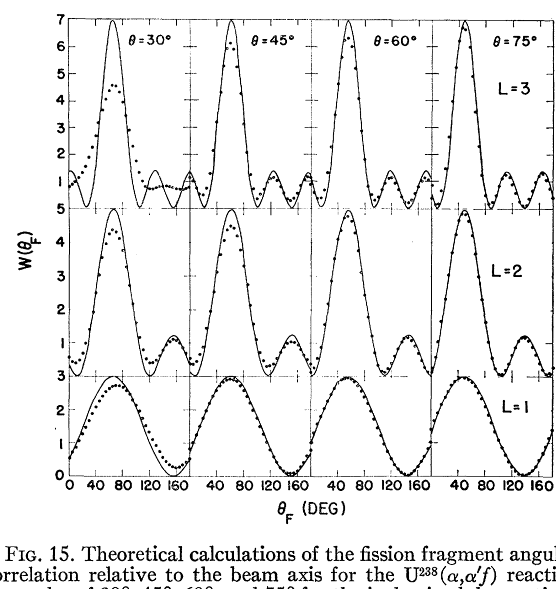

FIG. 15. Theoretical calculations of the fission fragment angular correlation relative to the beam axis for the $U^{238}(\alpha,\alpha' f)$ reaction for angles of 30°, 45°, 60°, and 75° for the inelastic alpha particle. Solid curves show the results of plane-wave calculations. The results from DWBA calculations are indicated by solid circles.

[19] R. H. Bassel, R. M. Drisko, and G. R. Satchler, Oak Ridge National Laboratory Report No. ORNL-3240, UC-34-Physics, 1962 (unpublished).

[20] G. Igo, Phys. Rev. **115**, 1665 (1959).

and is consistent with the experimental results which gave a difference of $4°\pm 2°$ between the symmetry angle and the kinematic recoil angle. Calculations were also performed for $3<l\leq 10$ but in these cases Eq. (A1) was summed for terms $0\leq L\leq 6$. Even though the complete angular correlation was not calculated for these cases it was still possible to compare plane-wave and DWBA results. These calculations indicated that for $\theta_\alpha=75°$ the DWBA results were also essentially the same as the plane-wave predictions for $3<l\leq 10$. These results are interpreted as justification for the use of a plane-wave analysis of the angular correlations for $\theta_\alpha=75°$.

In addition to the angular correlations the DWBA calculations were also used to determine the relative cross sections as a function of the l transfer. The results show that $\sigma(l)/(2l+1)$ is approximately independent of l through $l=9$ and then starts to decrease at $l=10$. In analyzing the experimental results the approximation was made that $\sigma(l)=2l+1$ for $0\leq l\leq 10$ and $\sigma(l)=0$ for $l>10$.

APPENDIX II. REANALYSIS OF (*d,pf*) ANGULAR CORRELATIONS

The experimental angular correlations from the $Pu^{239}(d,pf)$ experiment[7] were found to be consistent with the form

$$W(\theta)\propto 1+\bar{g}_2P_2(\cos\theta). \qquad \text{(B1)}$$

In order to compare these results with the present $(\alpha,\alpha' f)$ results, it is necessary to calculate $\bar{g}_2$ for the various combinations of vibrational bands which appear to contribute to the $(\alpha,\alpha' f)$ angular correlations.

In performing these calculations the assumptions were the same as those made for the general development of the theory for (d,pf) reactions[7] except that it was assumed that, in the residual nucleus, states with $J\leq 7$ of either parity were equally available. Then the angular correlations for a specific angular momentum and parity $J\pi$ could be determined as follows: Each orbital angular-momentum transfer, l, populates two neutron orbitals, $j=l\pm\frac{1}{2}$. These states have a parity given by l (positive if l is even, negative if l is odd). The neutron orbitals $\frac{1}{2}\leq j\leq 15/2$ with both parities are populated from $0\leq l\leq 7$ and the relative population of a given state $j\pi$ can be obtained from the cross section for the appropriate l transfer, $\sigma(l)$. Values of $\sigma(l)$ are obtained from DWBA calculations.[21] Then the neutron angular momentum is coupled to the target spin ($I=\frac{1}{2}$ for Pu^{239}) to obtain the total angular momentum J for the fissioning nucleus. Each final state, $J\pi$, can be populated from two neutron orbitals $j=J\pm\frac{1}{2}$. Thus, for fission through a specific transition state, $JK\pi$, for $I=\frac{1}{2}$ the angular correlation is given by

$$g_2(JK\pi)=\frac{\sigma(j_+\pi)g_2(j_+JK)+\sigma(j_-\pi)g_2(j_-JK)}{\sigma(J_+\pi)+\sigma(j_-\pi)},$$

where

$$j_\pm=J\pm\tfrac{1}{2},$$

and

$$g_2(jJK)=\frac{(2L+1)(LJ0K|LJJK)(Lj0\tfrac{1}{2}|Ljj\tfrac{1}{2})^2}{(LJ00|LJJ0)}. \qquad \text{(B2)}$$

The relative cross section for forming a state $J\pi$ is given by

$$\sigma(J\pi)=\sigma(j_+\pi)+\sigma(j_-\pi). \qquad \text{(B3)}$$

Now it is possible to calculate a value of $\bar{g}_2$, the angular correlation averaged over fission through the various $JK\pi$ transition states which are available in a specific excitation energy interval. There are two models for the fission process which can be used to estimate $\bar{g}_2$.

(1) $\Gamma_f(J\pi)\approx\Gamma_T(J\pi)$.

This case is appropriate to fission at excitation energies below the neutron-binding energy. In this case fission competes only with gamma-ray de-excitation, and it is assumed that when a transition state of a specific $J\pi$ is available then all of the compound nuclei formed in states $J\pi$ will fission. This means that if several transition states with the same $J\pi$ but different K are available then the fission will be split equally between these states. Then for fission of specific states $J\pi$ the angular correlation is given by

$$g_2(J\pi)=\sum_i g_2(J\pi K_i)/N(J\pi), \qquad \text{(B4)}$$

where the sum is taken over all available transition states with a given $J\pi$ and $N(J\pi)$ is the number of these states available. The total angular correlation is then obtained by summing over all the states which can be formed

$$\bar{g}_2=\sum_{J\pi} g_2(J\pi)\sigma(J\pi)/\sum_{J\pi}\sigma(J\pi). \qquad \text{(B5)}$$

(2) $\Gamma_f(J\pi)\ll\Gamma_T$.

The alternative model for calculating $\bar{g}_2$ assumes that the probability of fission is small compared to other de-excitation modes. This model gives a lower limit for $\bar{g}_2$ in the region where neutron re-emission can compete with fission. In this case if the probability of fission through any transition state, $J\pi$, is proportional to the formation probability for that state, $\sigma(J\pi)$, then

$$\bar{g}_2=\sum_{J\pi} g_2(J\pi)N(J\pi)\sigma(J\pi)/\sum_{J\pi}N(J\pi)\sigma(J\pi). \qquad \text{(B6)}$$

Using Eq. (B5), calculations of $\bar{g}_2$ were performed assuming the presence of various vibrational bands in the excitation energy range up to ~1.6 MeV above threshold for $Pu^{239}(d,pf)$. For higher excitation energies where neutron re-emission is possible, Eqs. (B5) and (B6) give upper and lower limits for the expected experimental values. Results from these calculations are shown in Fig. 14.

[21] W. R. Gibbs (private communication).