Analyses of fast neutron inelastic scattering cross sections to higher (vibrational) states of ^{232}Th and ^{238}U. I. Standard formalism

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Angle-integrated cross-section data in the form of excitation functions from threshold to 2.5 MeV incident (lab) energy, as measured and calculated, are presented for inelastic scattering of fast neutrons on ^{232}Th and ^{238}U, proceeding to higher collective (quadrupole and octupole vibrational) states of these deformed actinide nuclei. The experimental (n,n') data were for the most part obtained from corrected γ-production $(n,n'\gamma)$ yields. The theoretical data were derived from an incoherent sum of the compound-nuclear and strong-coupling direct-interaction cross sections, computed using the Bruyères set of optical potential and deformation parameters. The relative coupling strength was the sole adjustable parameter. Provision was made for the effect of Moldauer level-width fluctuations and for competing neutron exit channels (but not for radiative capture, charged-particle, or fission competition); the coupled-channels computations permitted up to six levels to be coupled simultaneously. The ensuing "standard" theoretical level excitation functions matched the experimental data fairly closely in most instances, confirming the viability of this conventional method of analysis. However, indications of potentially still better fits were provided by preliminary calculations employing the unified statistical S-matrix formalism of Weidenmüller *et al.* As illustrated for (n,n') scattering to the lowest members of the $K=0^-$ octupole band in ^{232}Th and ^{238}U, the latter approach reproduces experimental trends closely when the relative coupling strength is optimized. A subsequent paper will present detailed results of the unified analyses and comparisons with evaluated neutron data-file excitation functions for these actinide target nuclei.

NUCLEAR REACTIONS ^{232}Th(n,n'), ^{238}U(n,n'), $E_n=0.8-2.5$ MeV; measured and calculated $\sigma(E_n)$. Enriched targets; measured $(n,n'\gamma)$ γ production: deduced $\sigma(n,n')$. Calculations with statistical compound nucleus and coupled-channels direct-interaction formalisms. Deduced relative coupling strengths. Preliminary unified statistical S-matrix calculations.

I. INTRODUCTORY

The dynamics of energy transfer and the modulation of neutron energy flux in fission reactors are, among other physical factors, crucially determined by the neutron scattering processes that occur in the reactor core and surroundings. A detailed quantitative understanding of neutron scattering interactions with the principal actinide nuclei is therefore basic to the elucidation of the physics and nuclear engineering energetics of reactor design and operation. Experimental and theoretical research, particularly during the past decade, by a number of groups [principally, those at Lowell, Argonne, Oak Ridge, Bruyères, Harwell, and Southern Universities Nuclear Institute (SUNI-RSA)] has contributed to the steady progress in the refinement of the techniques and the improvement in the accuracy and reliability of the data sets, and their interpretation. This has been particularly evident in the acquisition of detailed information on the main actinide fuel nuclides in breeder reactor technology, ^{232}Th and ^{238}U. The body of data for neutron interactions with these nuclei has been gathered in successive Evaluated Nuclear Data File (ENDF) evaluations, the most recent being the ENDF/B-V listings.[1,2] With each improvement in precision, the discrepancy between measured data and the successive

TABLE I. Deformed optical potential parameters for neutrons on actinide nuclei (Th, U, and Pu).

Nucleus	Energy E_n (MeV)	V_0 (MeV)	r_0 (fm)	a (fm)	W_{WSD} (MeV)	r_0' (fm)	a' (fm)	V_{so} (MeV)	$(r_0)_{so}$ (fm)	a_{so} (fm)	β_2	β_4	Ref.
^{232}Th	0.7–3.4	$46.4-0.3E_n$	1.26	0.63	$3.6+0.4E_n$	1.26	0.52	6.2	1.12	0.47	0.190	0.071	8
^{235}U	0.7–3.4	$46.4-0.3E_n$	1.26	0.63	$3.3+0.4E_n$	1.26	0.52	6.2	1.12	0.47	0.220	0.080	8
^{238}U	0.7–3.4	$46.2-0.3E_n$	1.26	0.63	$3.6+0.4E_n$	1.26	0.52	6.2	1.12	0.47	0.198	0.057	8
^{238}U	1.1	56.09	1.15	0.40	10.20	1.23	0.17	7.5	1.24	0.62			62
^{238}U	1.9	54.97	1.16	0.45	5.31	1.30	0.29	7.5	1.24	0.62			62
^{238}U	2.5	55.38	1.17	0.46	6.91	0.85	0.56	7.5	1.24	0.62			62
^{238}U	Composite	$66.3-6.8E_n$	$1.02+0.09E_n$ (Mean=1.21)	$0.29+0.09E_n$ (Mean=0.48)	$11.26-1.98E_n$	$1.49-0.19E_n$ (Mean=1.08)	$-0.36+0.42E_n$ (Mean=0.41)	7.5	1.24	0.62			62
^{239}Pu	0.7–3.4	$46.2-0.3E_n$	1.26	0.63	$3.6+0.4E_n$	1.26	0.52	6.2	1.12	0.47	0.220	0.070	8
^{242}Pu	0.7–3.4	$46.0-0.3E_n$	1.26	0.63	$3.5+0.4E_n$	1.26	0.52	6.2	1.12	0.47	0.204	0.051	8

ENDF evaluations has lessened. The aim of the present investigation has been to identify and reduce still further the remaining disagreement between experiment and theory in order to arrive at a definitive, detailed description of the scattering interaction probability for fast neutrons with the principal actinide nuclides, ^{232}Th and ^{238}U, over the important incident-energy region from threshold up to several MeV.

II. PREVIOUS STUDIES

Of the numerous studies undertaken hitherto, a fairly comprehensive summary as of late 1979 is to be found in the Proceedings of the Knoxville International Conference on Nuclear Cross Sections for Technology,[3] which features papers by the first four of the above groups[4–8] and references to the publications of the remaining groups. The Lowell group in particular has meanwhile, in a preliminary form, presented the results of extensive analyses in poster sessions at the 1980 Polish Summer School in Nuclear Physics,[9] featuring experimental findings which are currently being prepared for publication[10,11] and theoretical level excitation functions for vibrational states in quadrupole ($K=0^+$ and 2^+) and octupole ($K=0^-$, 1^-, 2^-, and 3^-) bands of these deformed nuclei, calculated from a combination of compound-nuclear (CN) and direct-interaction (DI) computer codes. The accord between measured and calculated data has meanwhile been improved further as a result of refinements in the analytic techniques; in this paper we present the results for the lowest 21 vibrational states in ^{232}Th and lowest 17 vibrational states in ^{238}U, which supplement the fits obtained by the Bruyères group[8] to the lowest rotational levels of these nuclei. For consistency, the same optical potential and deformation parameters as those derived by Haouat *et al.*,[8] and as listed in Table I, were employed throughout for the deformed Woods-Saxon derivative (surface absorption) potentials with spin-orbit coupling. Table I also lists the Bruyères potential and deformation parameters for other actinides, and the Lowell "best-fit" potential parameters.

III. EXPERIMENTAL

The level (n,n') cross sections were derived from $(n,n'\gamma)$ gamma-ray production yields, and in part corroborated by direct (n,n') measurements. The salient points of the experimental and data-

processing techniques have been described elsewhere[4,5,10–13]; results were obtained for altogether 75 gamma-ray transitions from 43 levels in ^{232}Th up to an excitation energy $E^*=1738.1$ keV, and for 45 gamma-ray transitions from 27 levels in ^{238}U up to $E^*=1561.4$ keV at incident neutron energies up to 2.2 MeV (laboratory). The incident neutron beam was almost perfectly monoenergetic, produced by directing a typically 9 μA proton beam from the Lowell 5.5-MV model CN (HVEC) Van de Graaff accelerator onto a solid tritium-containing target (approximately 100 keV thick for 2.3-MeV protons), wobbled and water-cooled to minimize deterioration. The neutrons from the ^{3}H$(p,n)^3$He reaction impinged on disc scattering samples placed 8.5 cm from the neutron source; the ^{232}Th scatterer was 100% isotopically pure: a disc 3.69 cm in diameter and 1.13 cm thick, and the ^{238}U scatterer was enriched to 99.8%, with approximately the same dimensions (3.8 cm diameter, 1.2 cm thickness). Prompt gamma-ray spectra were acquired at 125° to the incident neutron flux with a 40-cm^3 Ge(Li) detector positioned at 88 cm from the center of the scatterer, used in conjunction with a time-of-flight (TOF) system and a surrounding NaI(Tl) annulus to suppress Compton events. Shielding of these two detectors was accomplished by arranging structures of lead, and paraffin loaded with lithium carbonate, together with a copper shadow bar. To reduce the neutron flux falling on the gamma-ray detector, a paraffin block was placed between this Ge(Li) detector and the metal disc scatterer. The incident neutron flux was monitored during the data runs by a long counter, a fission chamber, and an auxiliary TOF system consisting of a plastic Pilot U scintillator mounted on a photomultiplier tube. The calibrated long counter and fission chamber were used to measure the 0° neutron fluence in a TOF system; the absolute efficiencies of these detectors were established through comparison with a recoil-proton telescope whose absolute efficiency was known, and using accurately calibrated gamma-ray sources of appropriate energy.

Two 4096-channel energy spectra were accumulated simultaneously in a multichannel analyzer: The first of these was the time-gated, energy-gated, Compton-suppressed spectrum that provided the accepted data; the other spectrum contained those gamma-ray events which fell in the time window but were rejected by the annulus. It was thereby possible to check the influence of the annulus in the registering of valid spectral lines in the gamma-ray data.

Combining the measured gamma-ray yields at 125°, near a zero of the second-order Legendre polynomial

$$[P_2(\cos 54.74°)=P_2(\cos 125.26°)=0]\ ,$$

with information on the neutron flux, it was possible to extract the absolute corrected gamma-ray production cross sections for each transition at each incident energy with the aid of appropriate computer codes, and then to convert these to the respective (n,n') level cross sections for the determination of angle-integrated excitation functions. In the above $(n,n'\gamma)$ technique, the program KATHY was used in conjunction with other correction codes, such as NEVES and PENHA, to derive differential γ-production cross sections and their statistical uncertainties, including the statistical uncertainty of the spectral peak counts and of the underlying background. Among the various corrections that were thus applied was one for dead time in the data acquisition system for gamma-ray yields, one for internal conversion in the sample, and one for variations in the angular distributions of the source neutrons incident upon the finite scatterer. Furthermore, the differential cross sections were corrected (up to second order) for the effects of neutron multiple scattering and for attenuation of neutrons and gamma rays. The arrangements for (n,n') measurements differed in minor details: As described by Beghian *et al.*,[13] the scatterer disc was somewhat thinner (3.82 cm diameter, 0.53 cm thick), at 10 cm from the neutron source, oriented so that its normal bisected the angle between the source-scatterer and scatterer-detector directions. Pilot U scintillators were used for neutron detection, each with TOF electronics, wherein a "walk correction" was used to improve the time resolution of the main TOF system, and counts were fed into a shared 8192-channel analyzer for data storage. To attain and maintain an energy resolution of better than $\sim$20 keV at neutron energies above 2 MeV the pulsed and bunched accelerator compression system and data acquisition procedures were monitored continuously,[14,15] so

(a) flight-time dispersions were kept small through the use of fast scintillators and photomultiplier tubes (RCA-8854 and RCA-C31024), the Mobley compression system[14] being operated under optimum conditions;

(b) flight path dispersions were minimized through the selection of a disc scatterer oriented optimally;

(c) neutron velocity dispersions were reduced through the choice of a thin metallic lithium target to generate neutrons via the ^{7}Li$(p,n)^7$Be reaction;

(d) correction codes IMBUI and GAVEA, as compiled by our group,[16] were used to determine the differential neutron cross section, corrected for multiple scattering to second order and for path-length dispersions.

The errors and uncertainties inherent in the (n,n') technique as applied under our conditions have been discussed by Beghian *et al.*[13]; those for the $(n,n'\gamma)$ technique have been detailed by Karatzas *et al.*[12] In the latter case, total uncertainties in the gamma-ray production cross sections were typically assessed at $\lesssim 10\%$ for the strong γ transitions and 20% for the weak ones. The net error estimate of $\leq 10\%$ ensued from a weighted combination of

(a) the statistical error in calculating the yield (generally $1-10\%$; in some exceptional instances of weak transitions ranging to as high as 40%);

(b) uncertainty in the calibration of the Ge(Li) detector efficiency ($4-6\%$);

(c) uncertainty in the incident neutron flux ($4-5\%$);

(d) uncertainty in the target source→scattering sample distance ($\simeq 1\%$); and

(e) error in the determination (from weighings) of the number of nuclei in the scatterer ($<1\%$).

In the derivation of the (n,n') angle-integrated level cross sections (and their error limits) the corrected 125° differential $(n,n'\gamma)$ cross sections corresponding to decay of a particular level (after subtraction of the respective production cross sections for transitions from higher levels that cascade through the state under consideration) were multiplied by a factor of 4π in accordance with the following reasoning: The angular distribution expression expressed as a Legendre-polynomial expansion,

$$\frac{d\sigma}{d\Omega} \equiv \sigma(\theta) = \sum_{\nu=0,2,4,\ldots} a_\nu P_\nu(\cos\theta)$$

$$\equiv a_0[1 + \sum_{\nu=2,4,\ldots} a_\nu^* P_\nu(\cos\theta)] , \qquad (1)$$

in terms of weighting coefficients a_ν (or normalized weighting coefficients a_ν^*) which rapidly decrease in magnitude with increasing ν and which vanish identically beyond a certain ν value determined by angular momentum selection rules, indicates that the zeroth-order coefficient a_0 is to a close approximation equal to the 125° differential cross section, since $P_2(\cos 125°) \simeq 0$ and a_4^*, a_6^*,... are either negligibly small or zero (for instance, for ^{232}Th at $E_n = 1.0$ MeV the mean value of the 4th-order normalized coefficient is $\overline{a_4^*} = 0.0617$ and at $E_n = 2.5$ MeV is $\overline{a_4^*} = -0.0119$; at $E_n = 1.0$ MeV, all values were small except those for the four highest levels out of 14, which had $a_4^* \simeq 0.4$ — omitting these, one finds that $\overline{a_4^*} = -0.0197$). In using the $(n,n'\gamma)$ technique to derive ^{232}Th(n,n') cross sections up to $E_n = 2.1$ MeV, McMurray *et al.*[17] found that in all but one out of 13 cases of excitation of levels up to $E^* = 1721$ keV, the inferred level cross sections were of smaller magnitude than those obtained from (n,n') measurements, the differences ranging from about 30% to factors of 3 or more. They attributed the discrepancies between the results from these two methods to the following:

(a) the existence of strong $E0$ transitions;

(b) the nonobservation of low-energy branching gamma rays due to internal conversion and gamma-ray attenuation; and

(c) the consequent inability to make adequate corrections for the feeding of levels by gamma-ray transitions proceeding from higher levels.

The assumption that effectively only zeroth-order (and second-order) terms are of significance in the Legendre distribution expansion is also inherent in the ^{238}U(n,n') production and scattering cross section determinations from $(n,n'\gamma)$ data at 125° by the Oak Ridge group.[7] It has been our conclusion that while some discrepancies exist between the (n,n') cross sections determined by direct measurement as against those derived from $(n,n'\gamma)$ data, the general extent of the differences is smaller than that indicated by the SUNI group.[17] Wherever a comparison has been possible, our numerical data for the gamma-ray production cross sections have been found to show a slightly higher trend than those measured by McMurray *et al.*,[17] which resulted in our derived (n,n') cross sections being perceptibly more nearly in accord with those obtained by direct (n,n') measurement for ^{232}Th scatterers. In the case of ^{238}U$(n,n'\gamma)$ our gamma-ray cross section data agree closely with those obtained by the Oak Ridge group[7,18] using a Ta target as the neutron source in the Oak Ridge Electron Linear Accelerator (ORELA) facility. In deriving (n,n') level cross sections from their γ-production data, the Oak Ridge group's procedure incorporated a correction for level feeding by unobserved transitions from higher-lying states using hitherto unpublished branching ratios.[19] This caused the cross sections beyond $E_n = 1.5$ MeV to be lowered significantly, enhancing the extent of their conformity with the ENDF/B-V evaluation. Even so, significant discrepancies remain between measured and evaluated inelastic scattering data.

Some of the inherent discrepancies can be ascribed to the mode of presentation of the ENDF evaluation. In order to conserve file-storage capacity, the format combines data for groups of levels, thus obscuring the individual comparisons. Moreover, the prime consideration in arriving at the ENDF data is devoted to securing close fits to the relatively large total-neutron and elastic-neutron cross sections over the entire energy range, leaving the comparatively small, but in practice vitally important, inelastic-neutron scattering cross sections to be extracted by subtraction. The consequent sacrifice in the goodness of fit to nonelastic cross sections is evinced in the manner in which the evaluated "grouped" inelastic cross section generally drops steeply in magnitude with rising incident energy beyond a fairly narrow peak, while the measured or calculated magnitudes maintain or increase their value with increasing energy, due to the ever-growing direct inelastic scattering component. Such a sustaining of magnitude in the inelastic data would entail some adjustment to the elastic evaluation that would result in a somewhat worsened fit, but would more realistically reflect the actual findings. Explicit comparisons will be presented in a subsequent paper.

In summary, the experimental data acquisition and processing techniques appear to have attained sufficient accuracy in the case of the principal actinide nuclei, ^{232}Th and ^{238}U, to meet specifications in data users' request lists.[20–22] The precision is adequate to serve as a reliable basis for data analyses, level by level, over an appreciable range of energy. In the case of cross sections extracted from $(n,n'\gamma)$ yield measurements, as featured in the present paper, there are indications that preliminary and forthcoming direct (n,n') measurements, where feasible, can offer reassuring substantiation of their magnitudes and energy dependence, particularly when correction techniques still in the process of being explored more deeply and extensively[16,23–26] are applied. As preliminary analyses[9] yielded sufficiently close agreement with the experimental data to support a detailed, thoroughgoing study, an intensive program of analysis was carried through, as described in the following sections of this paper for the "standard" approach and in a subsequent paper (Paper II) for the "unified" S-matrix approach.

IV. THEORETICAL ANALYSES

Following the adoption of the conventional Bruyères optical potential[8] with the parameters listed in Table I, the complicated level schemes of the deformed actinide nuclei had to be decomposed into collective band families. The assignments depicted in Fig. 1 were derived (a) for ^{232}Th and (b) for ^{238}U, guided by the previous conjectures (Refs. 1, 2, 17, and 27–30) for these target nuclei and by the systematic collective dependence of excitation energy E^* upon spin J, as indicated either by the centrifugal-stretching model or by a rotational-vibrational interaction model[31] through the formula

$$E^*(J)=E^*(0)+A[J(J+1)]+B[J(J+1)]^2\,, \tag{2}$$

where A and B are adjustable band parameters whose best-fit values are tabulated in Tables II and III. In arriving at the schemes of Fig. 1 used in the following analyses the decision was made to allocate the trio of levels in ^{232}Th at 1183, 1218, and 1329 keV to a $K=3^-$ octupole band (rather than to leave them in the $K=1^-$ band, for which the parameters and E^* sequence would be nonregular) and in ^{238}U to assign the 927-, 966-, and 1055-keV trio of $K=0^+$ levels to the γ-vibrational two-phonon band (rather than, as others[2,29] have done, to a one-phonon β band or quasi-β band), in conformity with the findings of Hess *et.al.*,[30] thereby interchanging the respective $K=0^+$ γ-band and β-band multiplets. Since two $\lambda=2$ excitation phonons would be expected to have roughly the same energy as one $\lambda=3$ phonon, the lowest member of the $K=0^+$ two-phonon γ band (at $E^*=927$ keV) would be expected to lie at approximately the average of the lowest excitation energies of the $K=0^-$, 1^-, and 2^- one-phonon octupole bands (for which $\bar{E}^*=913$ keV); it is not unreasonable for the lowest $K=0^+$ γ-band level to be of slightly lesser excitation energy than the lowest member of the β band, even though this appears not to be the case in ^{232}Th (it will be noted, though, that for this nuclide also, the energy of the lowest $K=0^+$ γ-band state, at $E^*=1078$ keV, is close to the one-phonon octupole band's energy average, $\bar{E}^*=1007$ keV). Since any error in the assignment of these levels to any particular vibrational band has significant repercussions upon the cross section derived from coupled-channels calculations as employed in these analyses, the finding of agreement between the observed and calculated results constitutes strong substantiative evidence attesting to the validity of the level-scheme classification.

As is consistent with the finding[32,33] of the onset and generally rapid dominance of an appreciable direct-interaction component, only some 1–2 MeV above threshold for neutron scattering from these heavy deformed target nuclei, the "standard ap-

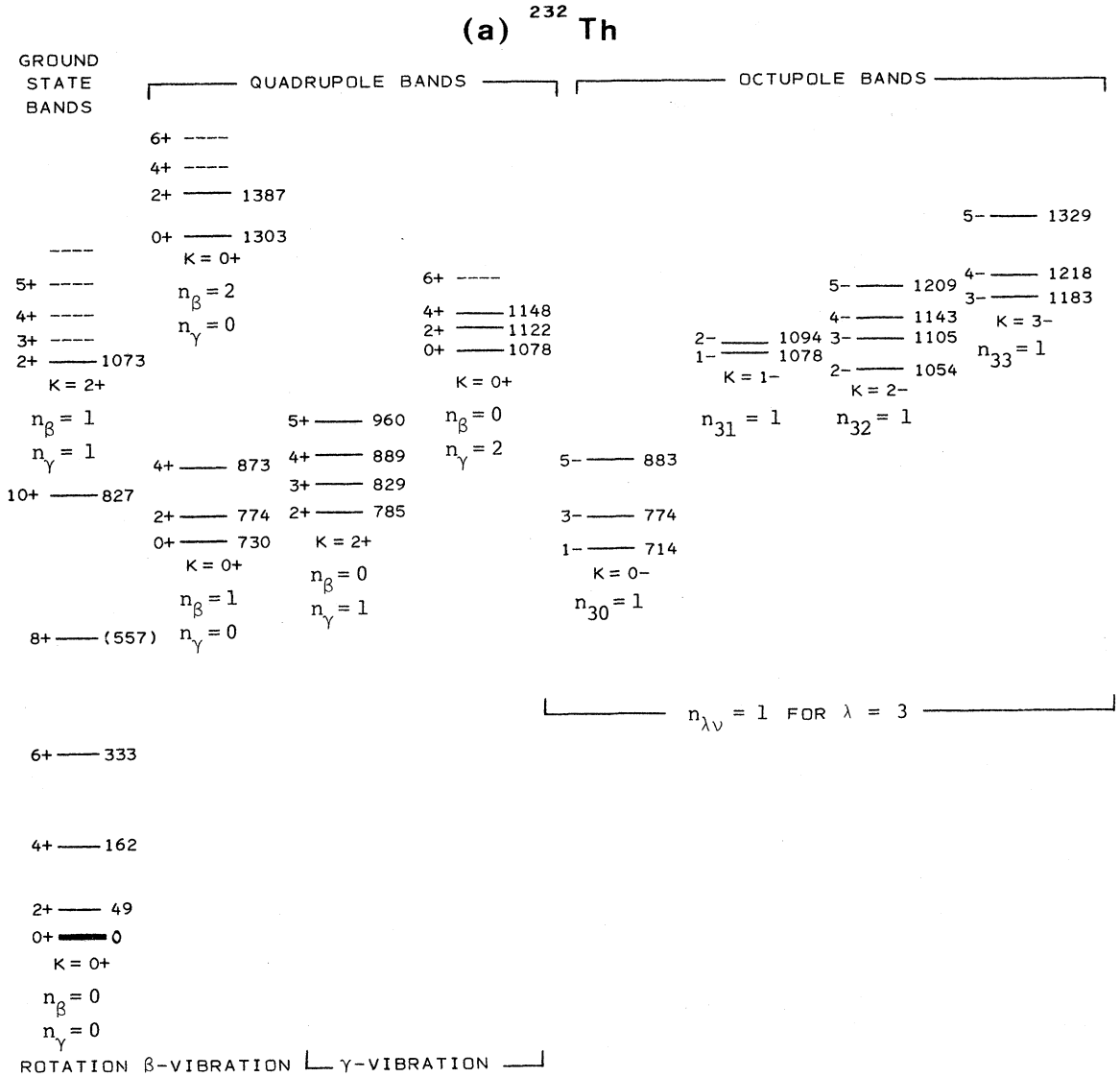

FIG. 1. (a) Decomposition of the collective (rotational and vibrational) band level scheme of ^{232}Th, based on compilers' suggested assigments (Refs. 1, 17, 27, and 29). A $K=3^-$ octupole band has been introduced in order to attain a regular progression of excitation energy with increasing spin J. (b) Corresponding collective band scheme for ^{238}U levels, based on the ^{232}Th scheme above, as well as on compilations (Refs. 28 and 29) and general rotational-vibrational calculations (Ref. 30). The levels in the $K=0^+$ bands are in conformity with these calculations (Ref. 30), but interchanged with those proposed by Sakai and Rester (Ref. 29) (see discussion in the text). The level excitation energies are those in Nuclear Data Sheets (Refs. 27 and 28) and differ slightly from those adopted in the experimental and theoretical studies by the Lowell group used in the present paper.

proach" of incoherently combining a compound-nucleus (CN) contribution with the corresponding direct-interaction (DI) contribution was followed in the analyses, using the computer programs CINDY[34] and JUPITOR,[35] or the variant KARJUP[36] prepared at Karlsruhe, to calculate the respective cross sections with the Bruyères set of optical-potential and deformation parameters employed throughout for consistency. Since the Moldauer level-width fluctuation correction was included in the determination of these parameters, provision for its effect was also incorporated in the CINDY calculations, which were based upon the customary Wolfenstein-Hauser-Feshbach-Moldauer statistical compound nucleus theory. Allowance was made for the influence of competing neutron exit channels (up to 55 of these

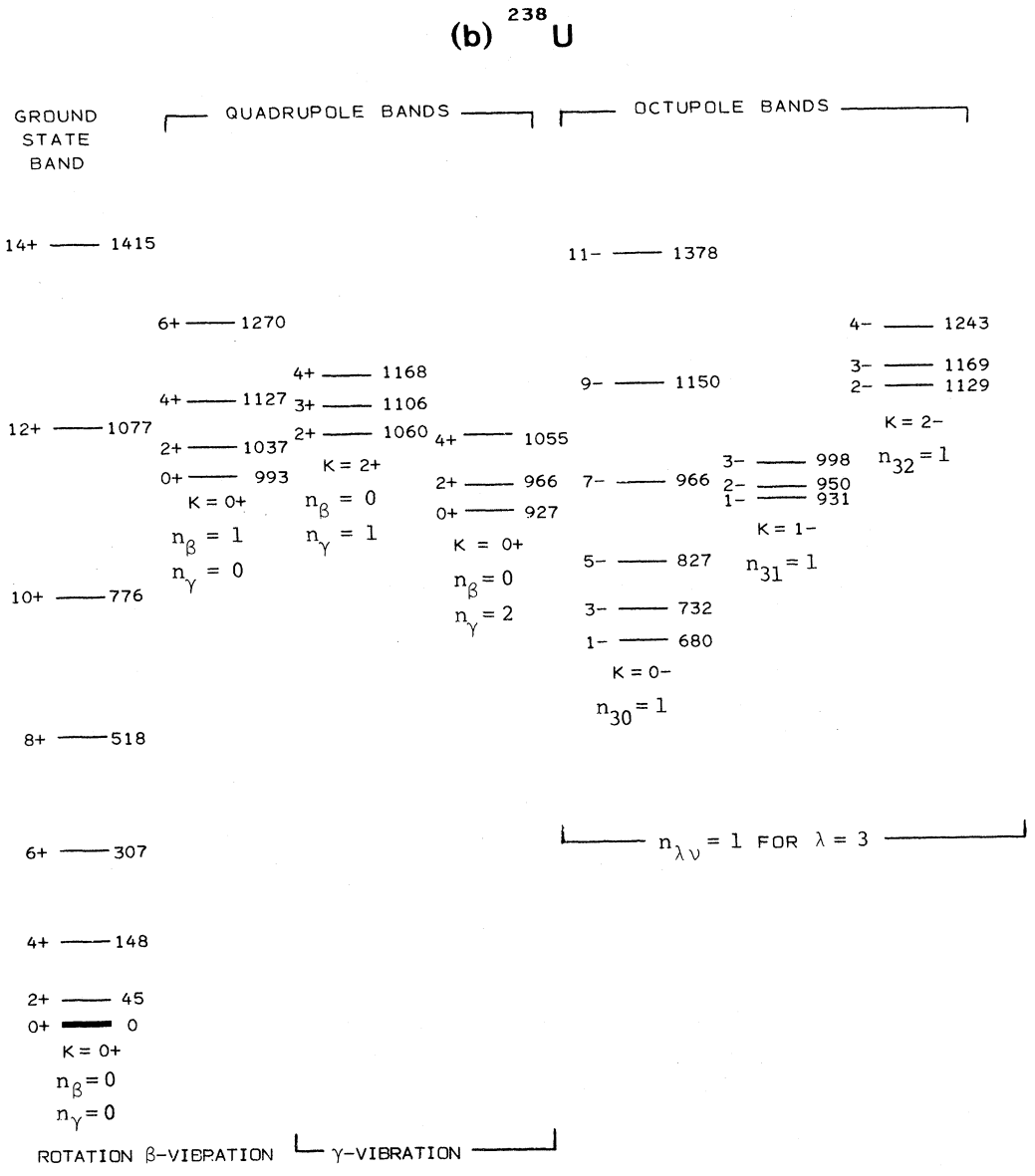

FIG. 1. (*Continued.*)

in the case of ^{232}Th, and 51 in the case of ^{238}U, at the highest incident energies). It was decided not to introduce a continuum artificially into the channel competition, even though the program has provision for this: In previous calculations[9] that contained continuum competition it was found that although close numerical agreement could be secured with numerical results obtained from such an approach by the Bruyères group,[37] the fit to the experimental variation of cross section with incident energy was less satisfactory than that which ensued from calculations that featured the observed levels explicitly. Nor was allowance made for the competition from radiative capture channels, since independent calculations and other findings[37] indicated that at most only a $\lesssim 1\%$ correction was entailed for the conditions under consideration. Moreover, to avoid encumbering the CN calculations with additional complexity, no correction for fission channels (a 3% effect, according to Yehia *et al.*[37] in the case of ^{232}Th in the present energy range) was introduced; for further investigations, however, plans are underway to incorporate provision for this into the CN program CINDY.

TABLE II. ^{232}Th evaluated collective state band configuration.

Band multiplet		Band parameters[a]			Spins	Excitation energies	
K	Type	E_0 (keV)	A (keV)	B (eV)	J^π	E^*_{calc} (keV)	E^*_{exp} (keV)
0^+	g.s. rot.	0.0	8.365	−12.26	0^+	0.00	0.00
					2^+	49.69	49.75
					4^+	162.21	162.4
					6^+	329.33	334.0
					8^+	538.08	557.8
					10^+	770.81	828.1
					12^+	1005.18	1138.8
					14^+	1214.09	
0^+	β vib.	730.1	7.414	−13.45	0^+	730.10	730.10
					2^+	774.10	774.1
					4^+	873.00	873
					6^+	1017.76	
0^+	2β vib.	1303.5	14.0	0.0	0^+	1303.5	1303.5
					2^+	1387.5	1387.2
					4^+	1583.5	
2^+	γ vib.	741.6	7.191	11.61	2^+	785.16	785.2
					3^+	829.56	829.6
					4^+	890.06	890.1
					5^+	967.78	960.2
					6^+	1064.10	
0^+	2γ vib.	1078.7	9.009	−276.43[b]	0^+	1078.7	1078.7
					2^+	1122.8	1122.8
					4^+	1148.3	1148.3
0^-	oct.	702.3	5.99	1.43	1^-	714.30	714.25
					3^-	774.39	774.1
					5^-	883.29	883.3
					7^-	1042.22	
2^-	oct.	981.0	13.805	−284.52[b]	2^-	1053.60	1053.6
					3^-	1105.70	1105.7
					4^-	1143.30	1143.3

[a]Band parameters in the formula $E^*_J = E_0 + A[J(J+1)] + B[J(J+1)]^2$.
[b]The high negative magnitudes of B render these values suspect; the attempt to fit the members of the $K=1^-$ octupole band was unsuccessful (e.g., the lower $J=1^-$ and 2^- multiplet members could not be fitted with the parameters $E_0=1218.17$ keV, $A=-7.426$ keV, and $B=371$ eV, which fit the higher 4^-, 5^-, and 6^- multiplet members).

The statistical assumption of random-phased overlapping closely-spaced levels was justified in the excitation range under consideration: For example, a 1 MeV neutron impinging on a ^{232}Th target occasions the formation of the intermediate compound nucleus ^{233}Th at an excitation energy $E^*=5.7$ MeV. When this value, with appropriate shell and pairing parameters, is inserted in the high-energy level-density formula of Gilbert and Cameron,[38] the total density of states (irrespective of their nuclear spin J) is found to be of the order of 10^8 per MeV, while that of just spin-$\frac{1}{2}$ states is roughly 2×10^6 MeV^{-1}. Even at an excitation energy of only $E^*=1$ MeV, the Gilbert-Cameron (low-energy) formula indicates the density of states (for all J values combined) to be ~50 MeV^{-1} and at $E^*=2$ MeV to be ~600 MeV^{-1} for nuclei in this mass region; a tight packing of states which is reflected in the values of the respective S- and D-wave strength functions. The CN calculations, incorporating only the actually observed levels at higher excitation, are therefore likely to underestimate the competition and may well yield slightly high cross sections at the higher incident energies.

Notwithstanding the high density of levels, the CN mechanism relinquishes its dominance to DI in general within about 1–2 MeV of threshold. Because the actinide nuclei are fairly strongly de-

TABLE III. ^{238}U evaluated collective state band configuration.

Band multiplet		Band parameters[a]			Spins	Excitation energies	
K	Type	E_0 (keV)	A (keV)	B (eV)	J^π	E^*_{calc} (keV)	E^*_{exp} (keV)
0^+	g.s. rot.	0.0	7.510	−4.524	0^+	0.00	0.00
					2^+	44.89	44.9
					4^+	148.38	148.4
					6^+	307.41	307.2
					8^+	517.17	517.8
					10^+	771.14	775.7
					12^+	1061.03	1076.6
					14^+	1376.80	1415
					16^+	1706.68	1788
					18^+	2037.17	2191
					20^+	2352.99	2619
					22^+	2637.14	3067
0^+	β vib.	993.0	7.676	−48.8	0^+	993.00	993
					2^+	1037.30	1037.3
					4^+	1127.00	1127
					6^+	1229.31	1270
2^+	γ vib.	1014.4	7.559	5.952	2^+	1059.97	1060.3
					3^+	1105.97	1105.6
					4^+	1167.97	1167.7
0^+	2γ vib.	927.0	6.614	−10.714	0^+	927.00	927.0
					2^+	966.30	966.3
					4^+	1054.99	1055
0^-	oct.	669.8	5.123	4.087	1^-	680.06	680.1
					3^-	731.86	731.9
					5^-	827.17	827.2
					7^-	969.51	966
					9^-	1163.98	1150
					11^-	1417.25	1378
					13^-	1737.57	1649
					15^-	2134.75	1959
					17^-	2620.16	2306
					19^-	3206.75	2687
1^-	oct.	924.9	2.307	311.67	1^-	930.76	930.8
					2^-	949.96	950.0
					3^-	997.46	997.5
2^-	oct.	1101.18	3.514	178.87	2^-	1128.70	1128.7
					3^-	1169.10	1169.1
					4^-	1243.00	1243 ?
					5^-	1367.57	1375 ?

[a]Band parameters in the formula $E^*_J = E_0 + A[J(J+1)] + B[J(J+1)]^2$.

formed, a strong-coupling approach is indicated for the DI computations. For this reason, the program JUPITOR and its Karlsruhe variant KARJUP were selected for the present investigation, after they had been established to provide results compatible with those from other DI codes such as DWUCK.[39] The DI computations were carried through independently of the CN calculations and of the experimental data acquisition in order to avoid bias, with the same set of optical potential parameters and other variables used throughout for consistency. At an early stage of the analysis, the program KARJUP was used in its search mode to derive best-fit optical parameters for ^{238}U+n at incident energies of 1.1, 1.9, and 2.5 MeV as a basis for comparison with those proposed by the Bruyères group. They are listed in Table I for reference, but were subsequently relinquished in favor of the standard Bruyères parameters, as also tabulated in Table I. No further attempt was made to "fine tune" the parameters as a means of enhancing the goodness of fit artificially; the only variable parameter left in the DI calcu-

lations was the relative coupling strength of the individual vibrational bands used in the collective rotational-vibrational model. The fitting procedure thus provided a means of assessing the strength of the coupling for the first time in nuclei of this type.

To render this extensive program of coupled-channels DI computations feasible within the generous but limited allocations of running time on the Lowell CDC Cyber 71 facility, it was necessary to streamline and optimize the procedures, in the course of which considerable modifications were made to the source programs. They were converted to single precision and, in the subroutine POTENT of JUPITOR the incorrect specification BR(4) was emended to BR(3) in statement number |SN|287+7. In KARJUP the subroutine RAC7 for the computation of Wigner 3-j (essentially, Clebsch-Gordan) coefficients was replaced for greater accuracy and speed. Through the use of the INTYPE=6 control option, up to six levels could be coupled together collectively; in all instances the 0^+ ground state was one of these, and the next (2^+ and, in most instances, 4^+) members of the ground-state rotational band were included in the group. Through the combinations of such ensembles, varying the coupling strengths for each band appropriately, it was possible to accommodate all the observed states in the lowest vibrational bands in turn, e.g., in Tamura's classification scheme,[35] the members of the $K=0^+$ ground-state rotational band (proceeding only as high as the 4^+ level) coupled to one another and to the vibrational multiplets: $K=0^+$ (β band), $K=2^+$ (γ band), and four negative-parity octupole bands, viz., $K=0^-$ (B band), $K=1^-$, $K=2^-$ (G band), and $K=3^-$. The program also allowed for individual higher collective levels to be coupled singly to the 0^+ ground state. As it did not have provision for strong coupling for two-phonon states, such as the members of the $K=0^+$ upper-β band or the $K=0^+$ γ band, pairing with the 0^+ ground state provided the only recourse for these. No attempt was made to extend the calculations beyond the 1217.6-keV state in ^{232}Th (the 4^- member of the $K=3^-$ octupole band), or the 1269.4-keV (6^+, β vibrational) level in ^{238}U. Nor were any analyses undertaken for states having spin higher than $J=6$, since no experimental data were available for these.

V. RESULTS

The ensuing findings for neutron scattering from threshold to 2.5 MeV are shown in Figs. 2–4 for 21 ^{232}Th levels and in Figs. 5–7 for 17 ^{238}U levels. The dots with error bars represent the Lowell measured data, with statistical error limits. The solid curves depict the resultant "standard" theoretical level-excitation functions, derived from an incoherent sum of the CN contribution (dotted-dashed curves) and the DI admixture (broken curves) to the net total cross section. The actual computed values from which these curves were obtained for incident energies from 0.8 to 2.5 MeV are tabulated in a subsequent paper, for comparison with the results of "unified" computations and with the ENDF/B-V evaluation.

In Fig. 2, the data for the 774.1-keV (2^+, β-vibrational) and 774.4-keV (3^-, octupole) one-phonon levels have been combined in view of the practical impossibility of experimental discrimination. An interesting feature, discernible in the data for the 883.3-keV (5^-, octupole) level of ^{232}Th in Fig. 2 (upper right) and, to a lesser extent, for its counterpart 826.7-keV (5^-, octupole) level in the $K=0^-$ band of ^{238}U in Fig. 5 (upper left), is the unusually gradual steady rise of the experimental and, likewise, the theoretical data from threshold: The conservative S-shaped upward progression is appreciably less precipitous than is customary for this region. In Fig. 7 a single set of experimental data for the 1128.9-keV level has been compared with the theoretical curves corresponding to either a 4^+ assignment (as for the second member of the $K=0^+$ β-vibrational band) or to a 2^- assignment (as for the lowest member of the $K=2^-$ octupole band). Since the nuclear spin of this state was not known, the more convincing fit derived with a 2^- assignment argues for the latter classification.

Inspection of the results overall reveals that a successful match between theory and experiment can be achieved in roughly one-half of the cases, particularly with those involving fairly low excitation of the residual states populated by inelastic scattering. Of the 20 cases (involving 21 levels because of the merger of the 774.1- and 774.4-keV state data) studied for ^{232}Th in Figs. 2–4, nine may be deemed to constitute a close fit, a further three evince too low a theoretical magnitude and eight too large a theoretical cross section over the entire incident energy range. In three instances (entailing 2^-, 4^+, and 4^- states) the experimental cross sections were too unreasonably small to be capable of being fitted by any theoretical calculation, no matter what the choice of approach or parameters. Only if the CN contribution were totally excluded could the experimental data be brought into accord with (DI) theory. Since no plausible reason can be

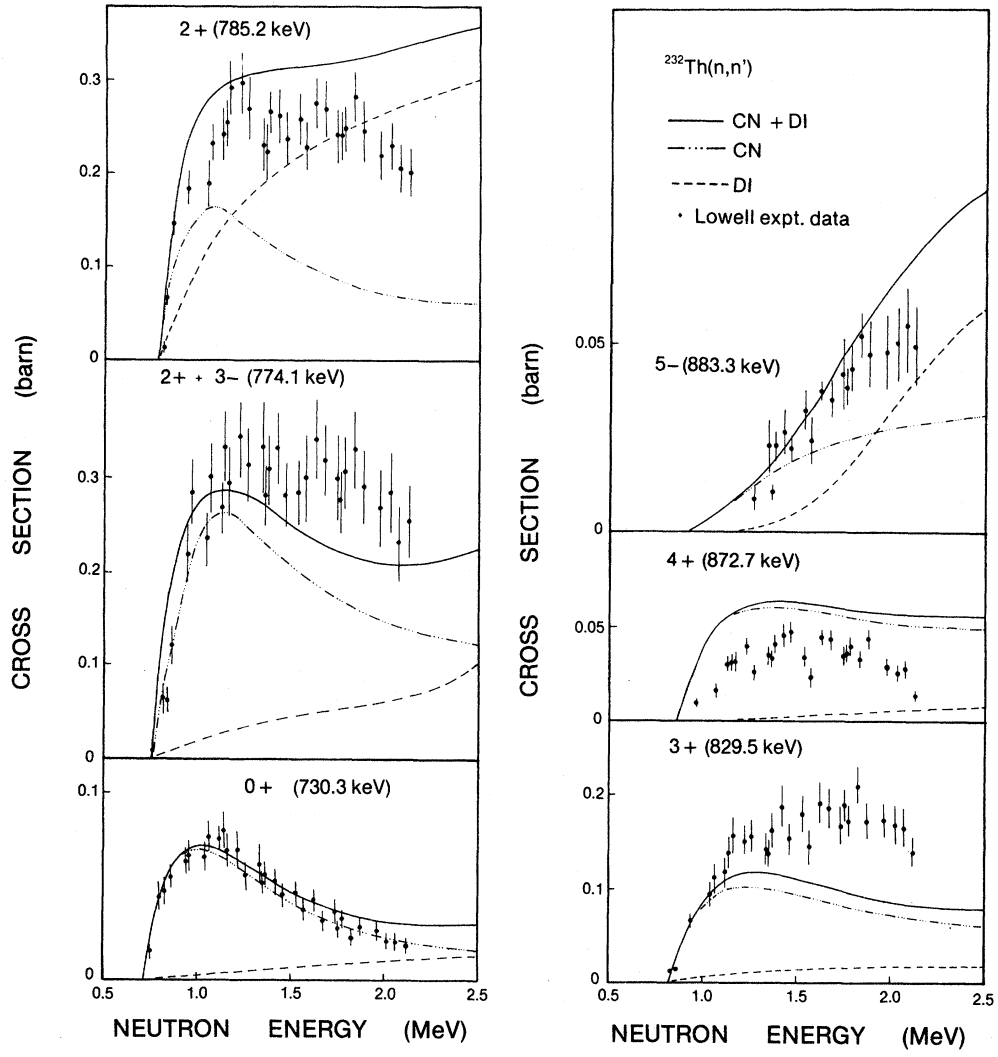

FIG. 2. Excitation functions for ^{232}Th(n,n') inelastic scattering to levels between 730.3 and 883.3 keV excitation energy (members of the $K=0^+$ β-vibrational, $K=2^+$ γ-vibrational, and $K=0^-$ octupole bands). The central left-hand figure refers to combined data for two indistinguishable states, viz., the 774.1 keV (2^+, β-vibrational) and 774.4 keV (3^-, octupole) levels. Experimental data (dots, with statistical error bars) are compared with standard (CN + DI, solid) theoretical curves, derived from incoherent summation of compound-nuclear (CN, dotted-dashed) and direct-interaction (DI, broken) contributions. The latter were obtained from coupled-equations computations; their magnitudes indicate that the DI admixture is appreciable, and in several instances rapidly equals or surpasses the CN cross section. The 3^- and 5^- level calculated cross sections are compared in Fig. 8 with those derived from a unified analysis based on Weidenmüller statistical S-matrix theory. The numerical magnitudes from which the theoretical curves were constructed are tabulated in a subsequent paper (paper II of our analytic treatment).

adduced for such an arbitrary exclusion, it may be argued that the fault lies with the experimental data here, due perhaps to wrong treatment of γ cascading and/or internal conversion in the derivation of (n,n') absolute cross sections from $(n,n'\gamma)$ measured yields.

In the case of ^{238}U, Figs. 5—7 reveal a further three such instances of anomalously low experimental cross sections (entailing 5^-, 4^+, and 4^- states) among the seven cases that evinced too high a theoretical cross section to be in accord with measured data. Perhaps the reason for this discrepancy is analogous to that above. Although the inclusion of additional competition channels in the CN calcu-

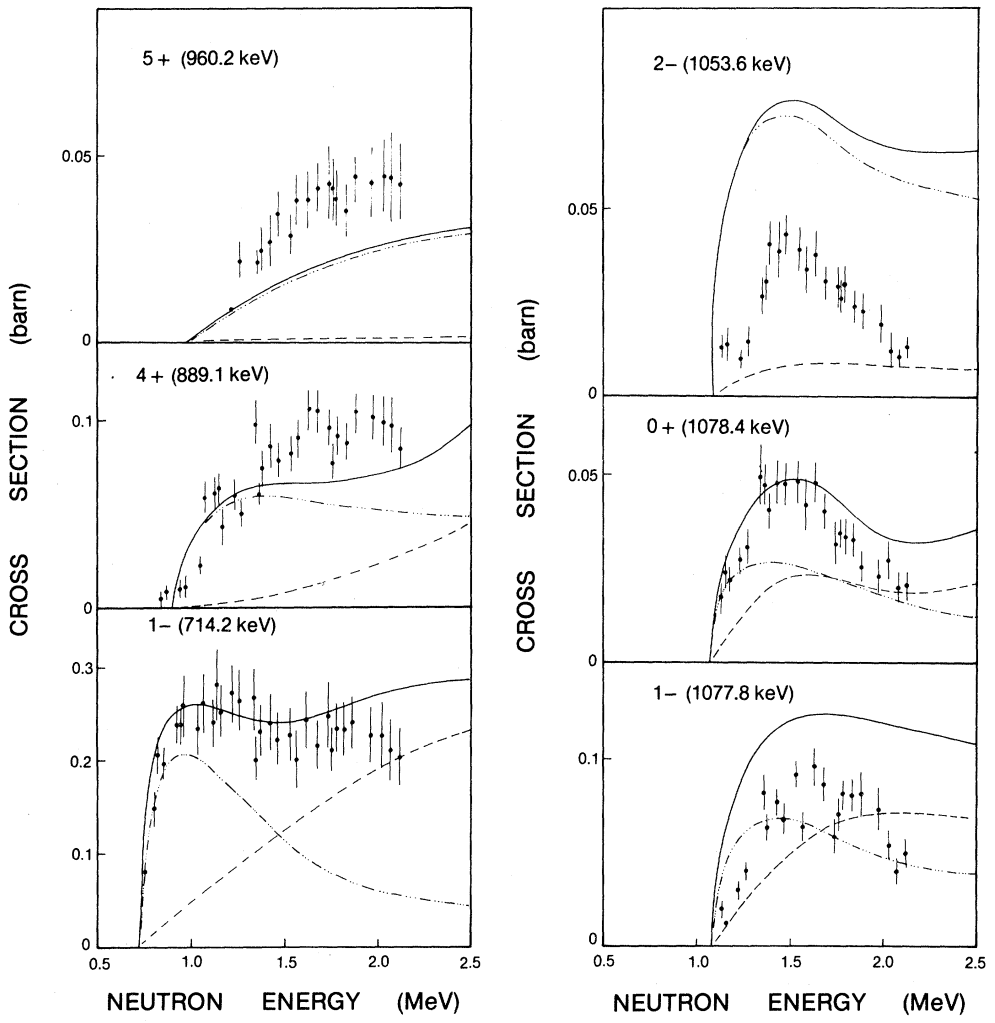

FIG. 3. Same as Fig. 2, but for ^{232}Th levels between 714.2 and 1078.4 keV that are members of the $K=0^-$, 1^-, and 2^- octupole bands, and of the $K=2^+$ and 0^+ γ-vibrational bands.

lations, including a fission channel, would have served to reduce the theoretical cross section further, the effect would be insufficient to account for the observed deviations. There were three instances of calculated cross sections being too low (namely, for 3^-, 4^+, and 6^+ states); only the first of these appears to be a genuine discrepancy, the second arose through the trial assignment of 4^+ to the 1128.9-keV level (fitted well with the 2^- assignment), which can be rejected, and the last featured a high-spin state for which the measurements have rather large errors and the calculated DI cross section is vanishingly low when derived with the same value of the coupling strength as was used for the other members of the β-vibrational band. Although in several instances it would have been tempting to vary the coupling strength for individual members of a multiplet, particularly for the highest members of the band, this *ad hoc* expedient was avoided. The remaining eight fits for ^{238}U levels were found to be of quite satisfactory quality.

VI. QUANTITATIVE ANALYSES WITH A UNIFIED APPROACH

Within the framework of statistical theory, the Hauser-Feshbach approach to the derivation of CN cross sections has, especially during the past decade, received ever deeper substantiation, at the basic level of theory as well as in practical application. Mol-

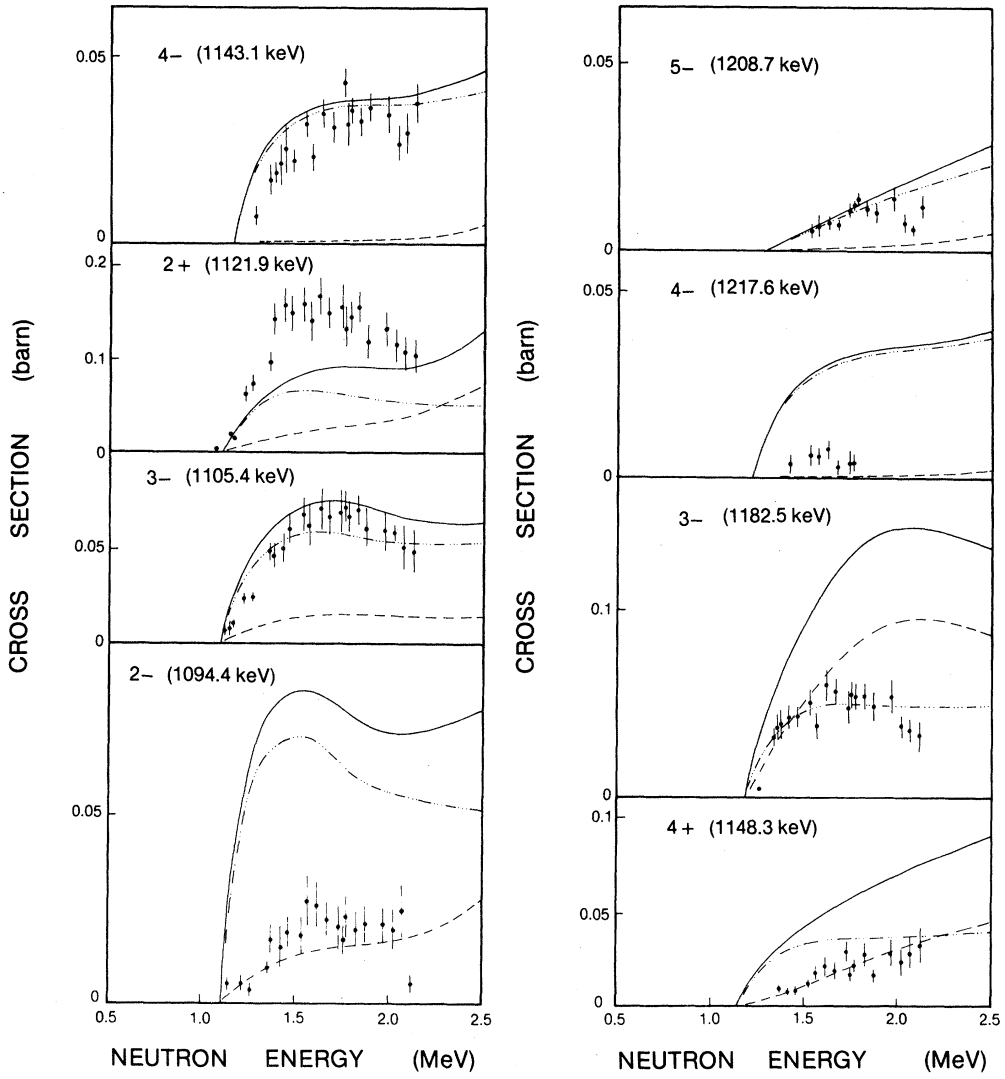

FIG. 4. Same as Fig. 2, but for ^{232}Th levels between 1094.4 and 1217.6 keV excitation energy that are members of the $K=1^-$ ($n_{\lambda=3,\nu=1}=1$), $K=2^-$ ($n_{\lambda=3,\nu=2}=1$), and the $K=3^-$ ($n_{\lambda=3,\nu=3}=1$) one-phonon octupole bands, as well as of the $K=0^+$ γ-vibrational two-phonon band. For these highly excited states, the fits are poorer in all but three cases, due possibly to erroneous corrections applied to the measured data through incorrect assumptions as to γ-cascade transitions and/or internal conversion.

dauer, in particular, has elucidated the necessary conditions for its validity, and adapted it to take account of level-width fluctuations and various statistical correlations. In his examination[39] of "why the Hauser-Feshbach formula works" he established the need for "M cancellation" as a fundamental requirement for the applicability of such a treatment to within a high degree of approximation. As a consequence of the unitarity and symmetry of the S matrix that embodies the actual physics of the interaction process, the channel-channel correlation effect brings about the cancellation of the influence of resonance interference terms,[40] $M_{cc'}$, and reduces the respective formulas[41,42] to well-defined, simple expressions amenable to straightforward numerical evaluation. The studies of Agassi *et al.*[43] in the limit of strongly overlapping resonances, and their recent extension and generalization by Hussein *et al.*,[44] have greatly assisted in placing this approach upon a sound, well-justified theoretical footing.

In the course of these investigations, the relevance of fluctuation analysis to the extraction of a comprehensive formalism that allows for the pres-

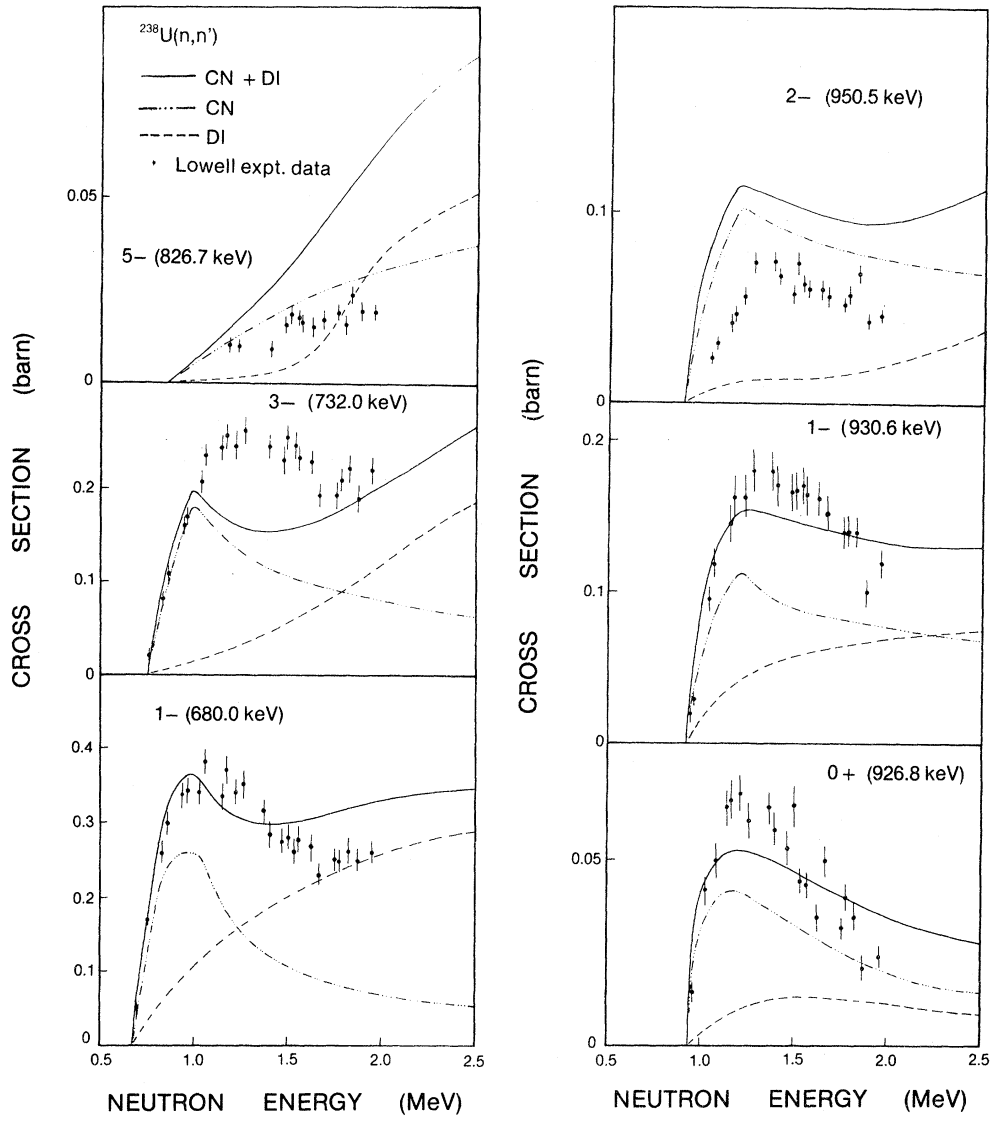

FIG. 5. Excitation functions for ^{238}U(n,n') inelastic scattering to levels between 680.0 and 950.5 keV excitation energy that are members of the $K=0^-$ and 1^- octupole band, and of the $K=0^+$ band [which we interpret as a γ-vibrational band, in conformity with calculations (Ref. 30)]. Again, experimental data derived from $(n,n'\gamma)$ measurements by the Lowell group (dots) are compared with standard (CN + DI) theoretical computations [solid curves, representing summed contributions from the CN mechanism (dotted-dashed curves) and the DI mechanism (broken curves)]; the numerical data from which the curves have been drawn are tabulated in a subsequent paper (paper II).

ence of direct reactions has come to the fore. General consistency conditions, derived by Engelbrecht and Weidenmüller[45] even in the presence of direct interactions from general properties of the statistical S matrix, had important repercussions upon the characteristics of resonance-parameter distributions and cross-section derivations. Specifically, the Engelbrecht-Weidenmüller transformation, diagonalizing the energy-averaged statistical S matrix ($\overline{\mathcal{S}}$) with the help of unitary matrix diagonalization procedures ($\mathcal{U}\overline{\mathcal{S}}\mathcal{U}^T$, where $\mathcal{U}^T$ is the transposed unitary $\mathcal{U}$ matrix), provided a means toward establishing a "unified, extended Hauser-Feshbach" theory in which the influence of direct interactions upon the (compound) fluctuation parameters is accommodated within a comprehensive formalism. The direct component, represented by the off-diagonal elements of the $\overline{S}$ matrix, can be treated

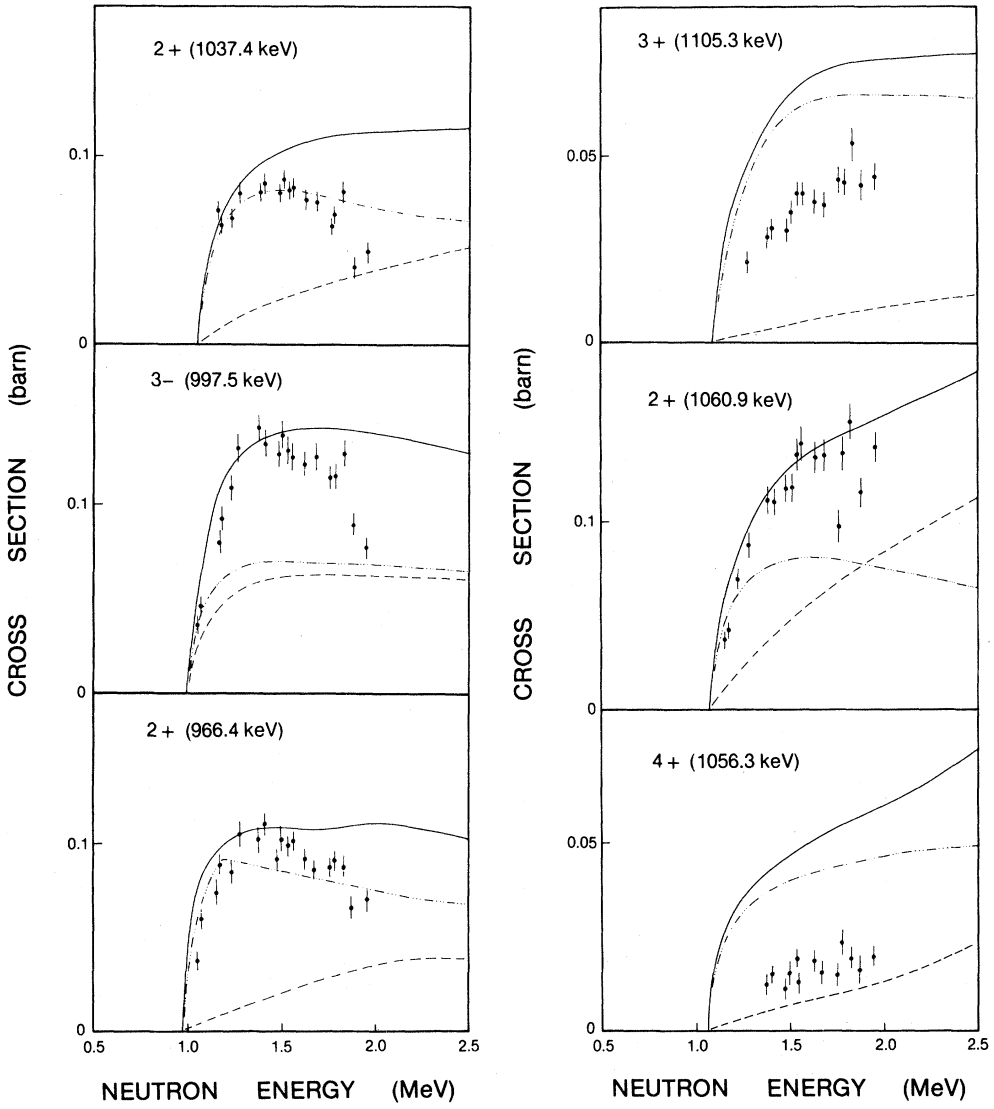

FIG. 6. Same as Fig. 5, but for ^{238}U levels between 966.4 and 1105.3 keV excitation energy that are members of the $K=0^+$ β-vibrational band (according to our assignment), of the $K=0^+$ γ-vibrational band (in our scheme), of the $K=2^+$ γ-vibrational band, and of the $K=1^-$ octupole band.

separately while the elements that are obtained from the diagonalization transformation can be used to build a fluctuation cross section. The net overall cross section can then be reconstructed from a combination of these contributions, wherein the energy averaging serves as a means of eliminating interferences between the components, since energy-averaged fluctuation amplitudes vanish statistically.

The detailed "unified" approach stemming from the statistical S matrix has been developed into a calculable formalism by Hofmann, Richert, Tepel, and Weidenmüller (HRTW).[46–48] This takes the mutual influence of all channels into consideration and allows for coupling between nuclear levels such that the influence of the direct coupling upon the fluctuation cross section is taken into account beyond just those effects which such direct couplings have upon the conventional transmission factors in the "standard" approach. The treatment thus lends itself naturally to the coupled-channels basis in, e.g., JUPITOR or KARJUP as used in the standard analyses described in the preceding, and invites comparison with the results obtained from the (CN + DI) approach.

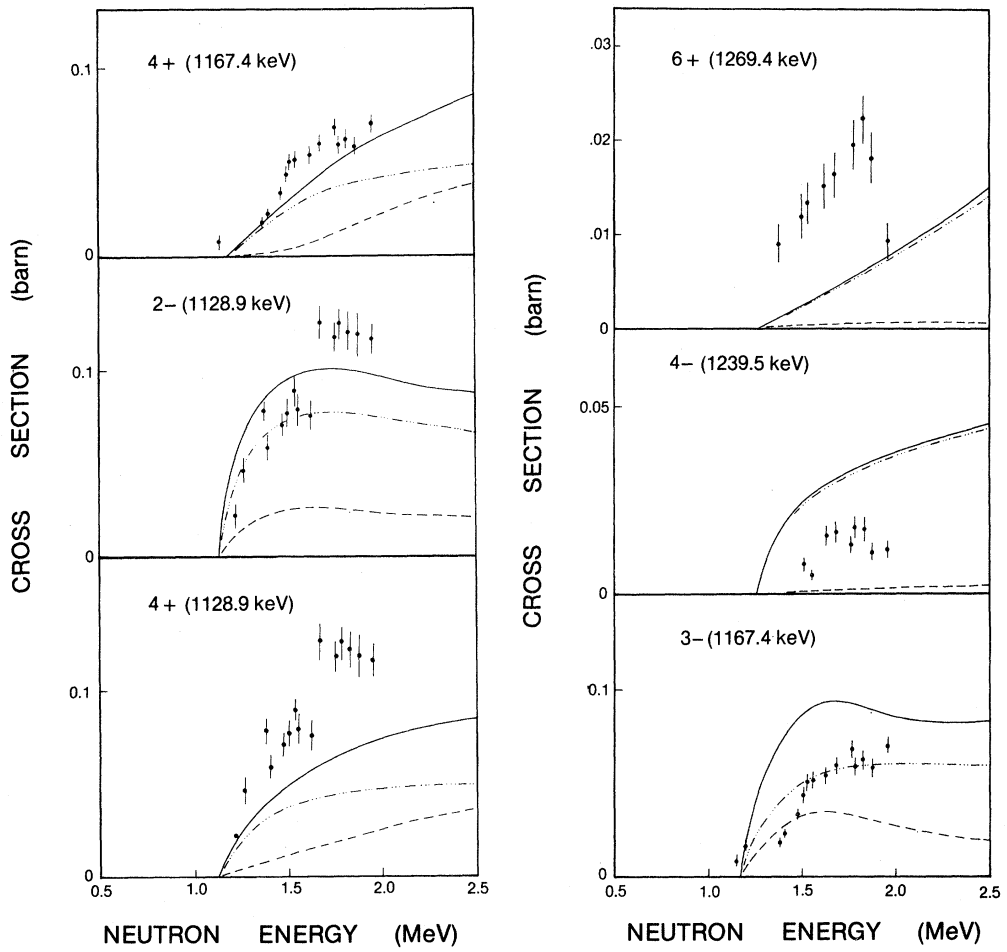

FIG. 7. Same as Fig. 5, but for ^{238}U levels between 1128.9 and 1269.4 keV excitation energy that are members of the $K=0^+$ (β-vibrational, in our scheme) band and of the $K=2^+$ γ-vibrational, as also of the $K=2^-$ octupole bands. A single set of data for the 1128.9-keV state has been analyzed separately in terms of a 4^+ and a 2^- spin assignment; the latter choice, corresponding to the lowest member of a $K=2^-$ octupole band, provides the more convincing fit.

Because of its numerical complexity, the statistical S-matrix formalism has in the past been applied only to a few cases in which special simplifying circumstances applied. Random-matrix theory was used in the original exploration[47,48] of its features to generate the extensive set of elements that compose the grand-ensemble matrices. Resonance isoanalog reactions were studied[49,50] with a simplified version of the Engelbrecht-Weidenmüller theory[45] to deduce resonance parameters in a situation in which only a very few strong compound channels were involved. Shortly thereafter, the analyses were repeated by Yoshida and Yazaki[51] for the isoanalog ^{92}Mo(p,p'_1) resonance reaction around $E_p=5.8-6.6$ MeV in which three fairly narrow compound analog states were featured, using more elaborate versions of the HRTW statistical S-matrix formalism to deduce the behavior of the time-development function, whose time integral yields the differential cross section. One of the versions is close to that obtained from the statistical theory of Kawai, Kerman, and McVoy[52] in which a number of questionable assumptions are made[39] in order to attain simplicity and which breaks down[53,54] when applied to the case of compound processes that compete with direct reactions. Another instance of application to analysis of isoanalog resonance data has been provided by the Erlangen group,[55,56] who studied the ^{90}Zr($\vec{p},p'\gamma$) reaction around $E_p=6-9$ MeV. The presence of four strong $T_>$ isoanalog states, serving as doorways for the neighboring $T_<$ compound states, and the absence of channel coupling, ren-

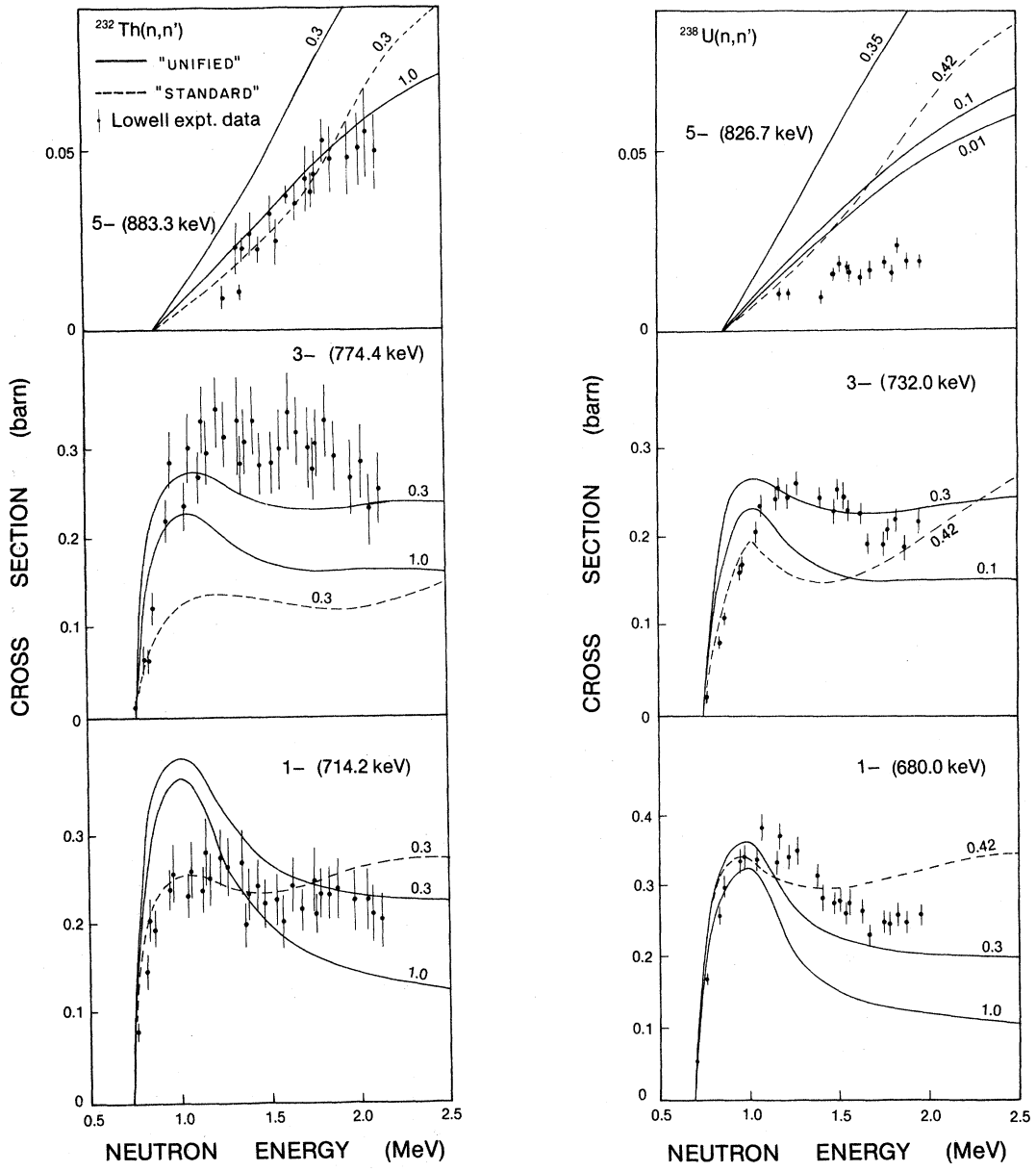

FIG. 8. Comparisons, for ^{232}Th(n,n') and ^{238}U(n,n') measured excitation function data (dots, with statistical error bars), of the predictions of unified statistical S-matrix theory (solid curves, for various values of the relative coupling strength as indicated beside the respective curves) and of standard (CN + DI) theory (broken curves, with "best-fit" values of the coupling strength as indicated). These preliminary analyses, applied to the members of the $K=0^-$ octupole vibrational bands in the residual actinide nuclei, suggest that the unified formalism is in principle able to provide a perceptibly better fit to experimental data than that attainable with the standard (CN + DI) approach. The detailed results of the unified analysis applied to the entire set of ^{232}Th and ^{238}U levels are presented in a subsequent paper (paper II), which also lists the numerical magnitudes from which all the theoretical curves have been assembled.

dered the calculations less complicated than they would otherwise have been within the HRTW framework. Similarly, the neglect of DI involvement in the HRTW calculations carried out by Konshin[57] to derive cross sections for neutron-induced reactions on ^{242}Pu and ^{239}Pu targets reduced the formalism to a variant that offered appreciable simplification, albeit at the expense of

eliminating information on possible channel-correlation interference and on feedback from direct amplitudes into the net cross section.

As applied now to an examination of ^{232}Th and ^{238}U(n,n') excitation-function data involving an intimate combination of CN and DI contributions and necessitating the consideration of channel coupling for collective levels, with many competing compound channels, the full content of the HRTW statistical S-matrix theory is drawn into scrutiny. For the evaluations, it was necessary to expand and adapt certain of the computer routines used in "standard" analyses and incorporate them in the composite program NANCY[58] designed to perform the elaborate, voluminous calculations. With the benefit of assistance from Moldauer,[59] the procedures for generating the complex S-matrix elements in JUPITOR or KARJUP (in which coupling is confined to only the ground state with each of ≤ 5 excited collective states) were extended and modified to permit simultaneous coupling of each channel with each of the other channels under consideration. Further competing neutron exit channels were accommodated through the inclusion of the penetrability routine SCAT[60] that served a similar purpose in CINDY (charged-particle channels were effectively precluded by the high Q values). For these competing channels SCAT had no provision for level-width fluctuation considerations, nor was it designed to deal with radiative capture or fission channels; these deficiencies are currently being remedied. The requisite energy averaging was accomplished automatically through the use of a complex optical potential (employing the same parameters as had been used in the "standard" calculations) that contained an energy-dependent imaginary absorption term. In ancillary investigations, it was established that this met the necessary averaging conditions without the need to invoke any subsidiary ensemble-averaging procedures. For the fluctuation treatment, the revised[47] formula expressing the "elastic enhancement factor"[46] was used as an adequate approximation, rather than its subsequently modified variant,[61] since this was the form presented in the original formulation of the HRTW theory. All other variables were retained numerically unchanged from the standard approach (e.g., deformations, competing channels, etc.); the only adjustable parameter was the relative band coupling strength. No attempt was thus made to "fine tune" any variables in order to procure a closer fit to experimental data. A specimen set of preliminary results is depicted in Fig. 8, which portrays data for the triplet of $K=0^-$ lowest octupole levels in the vibrational bands of ^{232}Th and ^{238}U, comparing the "unified" and "standard" findings against the experimental data of the Lowell group. The unified results compare favorably with those from the standard approach, and encourage the further exploration of the potentialities of the HRTW statistical theory, as pursued in paper II (see also Ref. 26, which tabulates the corresponding numerical results, as transmitted to the National Neutron Cross Section Center for archival).

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