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ACTINIDE NEUTRON ELASTIC SCATTERING DATA ANALYSES

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Abstract: Theoretical analyses are presented of experimental data for elastic fast-neutron scattering to the 0^+ ground states of the principal even-A actinides ^{232}Th, ^{238}U, ^{240,242,244}Pu, and to a "composite triad" of closely-adjacent ground-state levels of the odd-A nuclide ^{235}U in the form of "excitation" functions to 3.5 MeV and angular distributions at various incident energies up to 3.4 MeV. The measured data include the latest results and prior findings by the Lowell group. The analyses have been based on the "standard" (CN + DI) formalism, taking account of level-width fluctuations and extra exit-channel competition. These are contrasted with preliminary calculations in the statistical S-matrix (HRTW) formalism and with ENDF/B-V.

(neutron, elastic scattering, actinide, cross sections, excitation functions, angular distributions, compound nucleus, DWBA direct interactions, HRTW statistical S-matrix)

Introduction

As an adjunct to extensive prior studies[1-14] of fast-neutron *inelastic* scattering cross sections as a function of incident energy ("excitation functions") and scattering angle ("angular distributions") on the principal actinides, the Lowell group has now reverted[15,16] to investigating the *elastic*-scattering total (angle-integrated) and differential cross sections, taking competition from inelastic channels, radiative capture and fission into account, and making allowance for the effect of level-width (Moldauer) fluctuations.

Because of the closely-spaced and complicated structure of the respective level schemes of even-A ^{232}Th, ^{238}U, ^{240,242,244}Pu and the odd-A nuclide ^{235}U, as shown in Fig. 1 below (which depicts the onset of the lower vibrational levels interspersed among the rotational states, and demonstrates – particularly in the case of ^{235}U, presented with an expanded energy scale – the dense packing of mingled collective states), the experimental arrangements call for fine-discrimination techniques (as developed for the Lowell subnanosecond-resolution time-of-flight facility) and the theoretical approaches necessitate provision for the involvement of direct interactions coupling collective states in the scattering process.

The Lowell data, augmented where requisite[17] with that from other groups, are herein contrasted with theoretical excitation-function and angular-distribution curves obtained from computations of

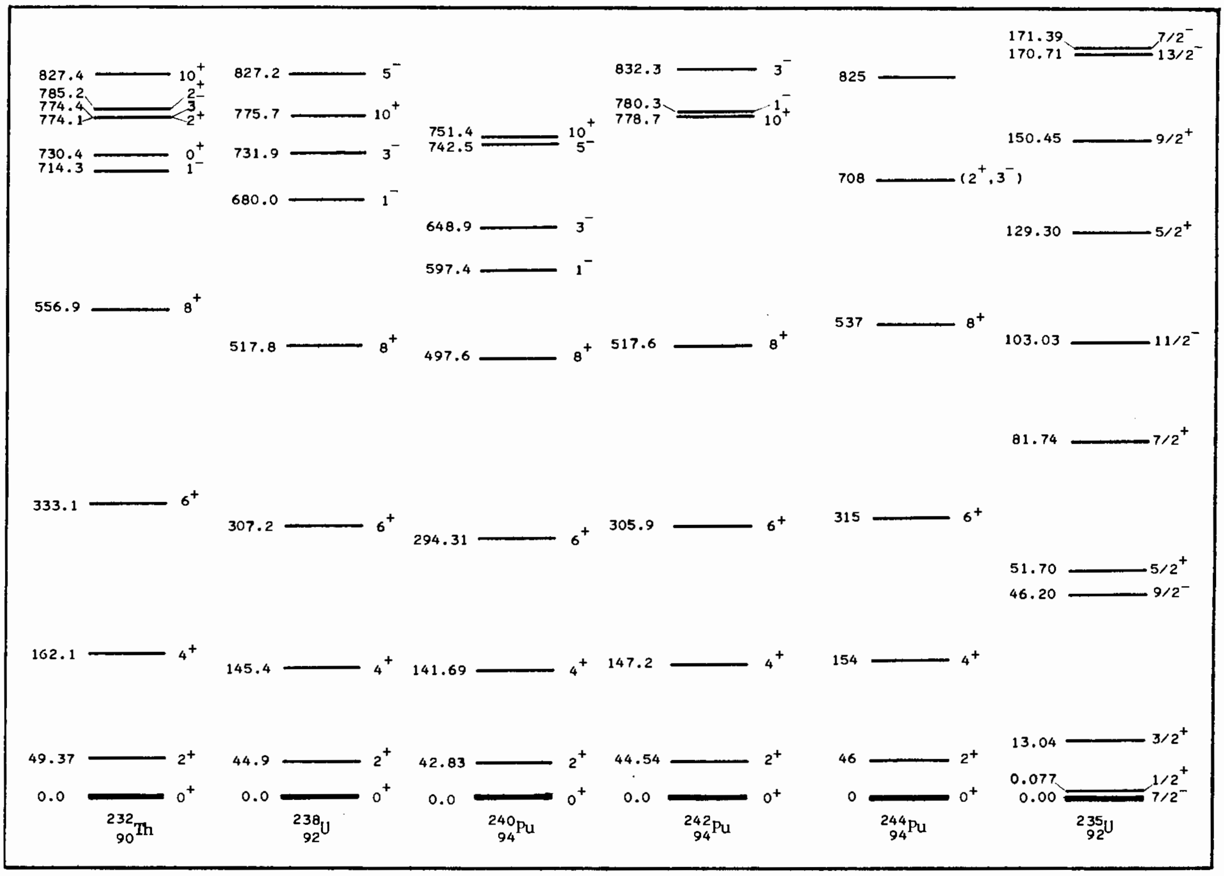


Fig. 1 Level schemes for the low-lying rotational and vibrational states (to ≃ 830 keV) of ^{232}Th, ^{238}U, ^{240,242,244}Pu and (to ≃ 170 keV, on an expanded energy scale) of ^{235}U.

total and differential cross sections with programs "CINDY" (CN, in Hauser-Feshbach-Moldauer formalism)[18] and "KARJUP" (DWBA coupled-channels formalism, the Karlsruhe variant[19] of Tamura's[20,21] code "JUPITOR") for the "standard" (CN + DI) approach *(solid curves)*, or with "NANCY" (HRTW, in Hofmann-Richert-Tepel-Weidenmüller statistical S-matrix formalism)[3,6,22-25] *(broken curves)*, and with evaluated ENDF/B-V data *(dotted curves)*.

Details of the computations are given in the next Section, with the main input parameters cited in the appended Table I. The elastic neutron excitation functions for the six actinides are presented below, in Figs. 2(a-c) for ^{232}Th, ^{235}U and ^{238}U, and 3(a-c) for ^{240,242,244}Pu. Thereafter, angular distributions at 185 keV for the first 3 nuclides are shown overleaf in Figs. 4(a-c) and at 550 keV in 5(a-c), with the remaining angular distributions and the concluding discussion on the final pages.

Computations

To preserve consistency, the optical model and deformation parameters (Table I) adopted by the Bruyères-le-Châtel group[26,27] were employed throughout the computations, as in the inelastic calculations heretofore.[9-14] The optical model was of the derviative (surface-absorption) Woods-Saxon type with a real Thomas-type spin-orbit potential and "global" parameters for neutrons on the actinides. Provision in CINDY was made for radiative-capture and continuum competition (as well as for inelastic n'-channels): the requisite level-density input parameters were those of Gilbert and Cameron.[28] For the (fertile) even-A actinides, fission competition was so slight as to be negligible over the entire energy range, but for the (fissile) odd-A nuclide ^{235}U a correction had to be applied to the output from CINDY in the form of a multiplicative factor

$$f \equiv (\sigma_{tot} - \sigma_f)/\sigma_{tot}$$

derived with the aid of output data from the statistical CN code "JACQUI" recompiled from Jacqueline Jary's Bruyères code[29] "NRLY". This provided fission cross sections σ_f and radiative capture cross sections σ_γ as well as total cross sections σ_{tot} (*i.e.*, the sum of shape-elastic, inelastic, fission and radiative-capture contributions) as a function of incident energy for each nuclide. Its output for ^{235}U yielded effective multiplicative correction factors f for CINDY data which ranged from 0.3245 at E = 0.1 MeV to 0.9918 at E = 2.5 MeV. By way of comparison, the commensurate values of f for ^{242}Pu ranged from 0.9985 to 0.9813. As the HRTW program NANCY presently lacks provision for radiative capture as well as fission channels, the appropriate correction factor multiplying the fluctuation (CN) cross section was modified to

$$f' \equiv (\sigma_{tot} - \sigma_f - \sigma_\gamma)/\sigma_{tot}$$

which, with data from JACQUI, ranged from 0.1684 to 0.9664 for ^{235}U over the energy range E = 0.1 - 2.5 MeV (while for ^{242}Pu the corresponding f'-values ranged from 0.9289 to 0.9617). As NANCY cannot take continuum levels into account, the cross sections that it provides are likely to be under-compensated, especially at the higher energies.

In the treatment of collective-level coupling by the programs KARJUP and NANCY it was convenient to follow the conventional practice for the even-A actinides of coupling only the rotational levels

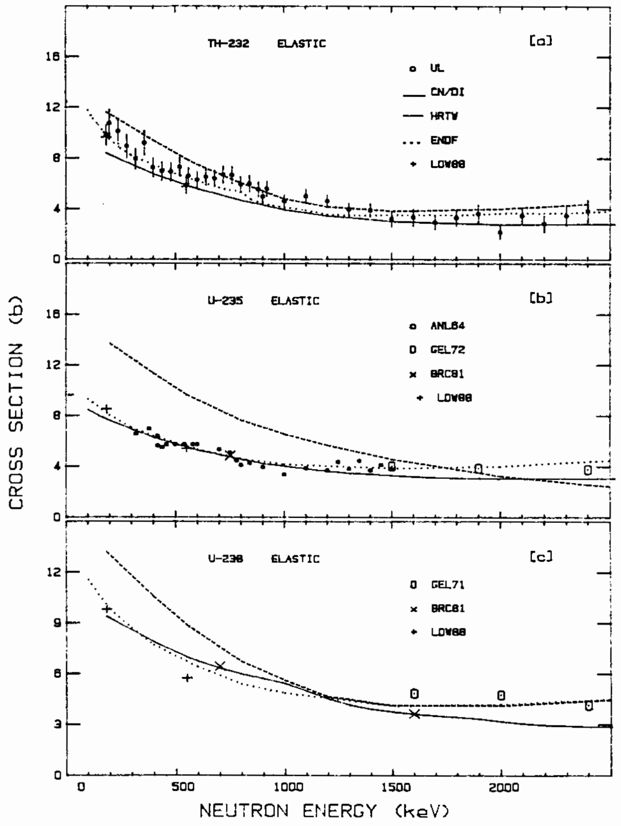


Fig. 2 Elastic neutron scattering excitation functions for (a) ^{232}Th, (b) composite ^{238}U, (c) ^{238}U to E = 2.5 MeV. Data[12,32,33,27,34] and curves are identified in the text.

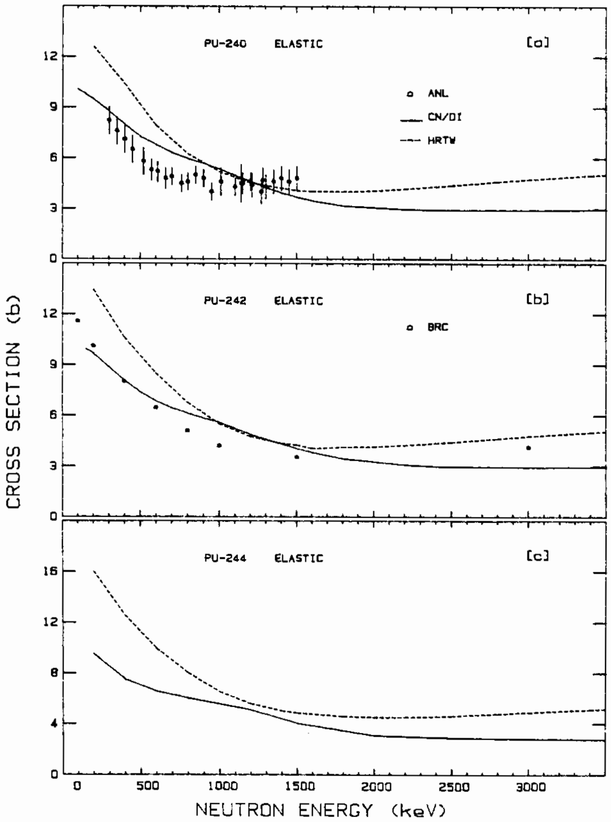


Fig. 3 Elastic neutron scattering excitation functions for (a) ^{240}Pu, (b) ^{242}Pu, (c) ^{244}Pu to E = 3.5 MeV. Data[35,27] and theoretical curves are identified in the text.

(with the option INTYPE set to 4) in the derivation of the elastic DI (nonfluctuation) cross sections. The alternative option (INTYPE = 2) of performing pure optical-model calculations devoid of any coupling to the 0^+ ground state was tried but rejected as it led to excessively high DI contributions. For the odd-A nuclide ^{235}U, special coupling provisions had to be made. Its triad of experimentally-unresolvable lowest-lying levels, $7/2^-(0)$, $1/2^+$ (77 eV) and $3/2^+$(13 keV) called for combination into a "composite ground state". This posed no difficulties for the calculations with CINDY in which the individual level contributions were simply combined into a net summed CN cross section, but for KARJUP and NANCY runs various expedients had to be tried out to find the most acceptable. The true $7/2^-$ ground state was unsuitable to serve as the "foundation" member of a ground-state band; instead, the $1/2^+$ (first-excited) level was adopted as an "ersatz" ground state upon which a $K = \frac{1}{2}^+$ rotational band was built (including the $3/2^+$ state as the next member), and to "patch in" the $7/2^-(0)$ level within the (INTYPE = 4) coupling scheme, specifying a sextet of coupled states in the input data: $1/2^+$(77 eV), $3/2^+$(13 keV), $5/2^+$(52 keV), $7/2^+$(82 keV), $7/2^-(0)$ and $9/2^-$(46 keV) *(see Fig.1)*. In the final analysis, it proved advantageous to run NANCY with the sequence $1/2^+$, $3/2^+$, $7/2^-$ and treat the remaining states (up to 37) as competition, progressively coupling 1, 2, and 3 states to get individual results for the $1/2^+$, $3/2^+$, $7/2^-$ cross sections *(dashed curves)* in Figs. 2, 4 and 5. Results from trial runs with KARJUP and NANCY using the alternative vibrational (INTYPE = 5) coupling option failed to provide convincing agreement. Although the present results from NANCY are preliminary and tentative, it seems reasonable to assume that the basic treatment is valid, when refined.

Findings

The elastic *excitation-function* results for the principal actinides are shown on the preceding page in Figs. 2 (to 2.5 MeV) and 3 (to 3.5 MeV). The ENDF/B-V evaluations *(dotted curves)*, designed to follow the data trends closely, do indeed provide a good match to the experimental data, which comprise measurements by the Lowell (UL & LOW88),[30,31] Argonne (ANL64),[32] Geel (GEL72),[33] Bruyères (BRC81),[27] Geel (GEL71),[34] and Argonne (ANL)[35] groups. The CN/DI (CINDY/KARJUP) computations *(solid curves)* likewise offer an encouragingly good fit to the data, while the HRTW statistical S-matrix (NANCY) results *(dashed curves)* lie generally rather high, particularly at the lower incident energies (where continuum competition is least).

In that *angular-distribution* analyses pose a yet more demanding challenge by entailing stringent comparisons of magnitude and structure, the results presented in Figs. 4 - 8 are especially noteworthy, particularly as they feature, conceivably for the first time, the application of the HRTW formalism in its full extent to the derivation of *differential* elastic cross sections as a function of angle. Figures 4(a-c) and 5(a-c) below contrast angular distributions at 185 keV and 550 keV for ^{232}Th and ^{235,238}U, wherein again the ENDF/B-V and CN/DI curves accord well with the data, while the HRTW calculations evince rather too pronounced a variation with angle. Since the NANCY results beyond 90° evoked misgivings, they have been suppressed in the plotted distributions, pending further scrutiny. It bears emphasizing that, as yet, these *differential HRTW findings are but preliminary and tentative*: the rough measure of agreement that they display is accordingly gratifying, yet a spur to further refinement.

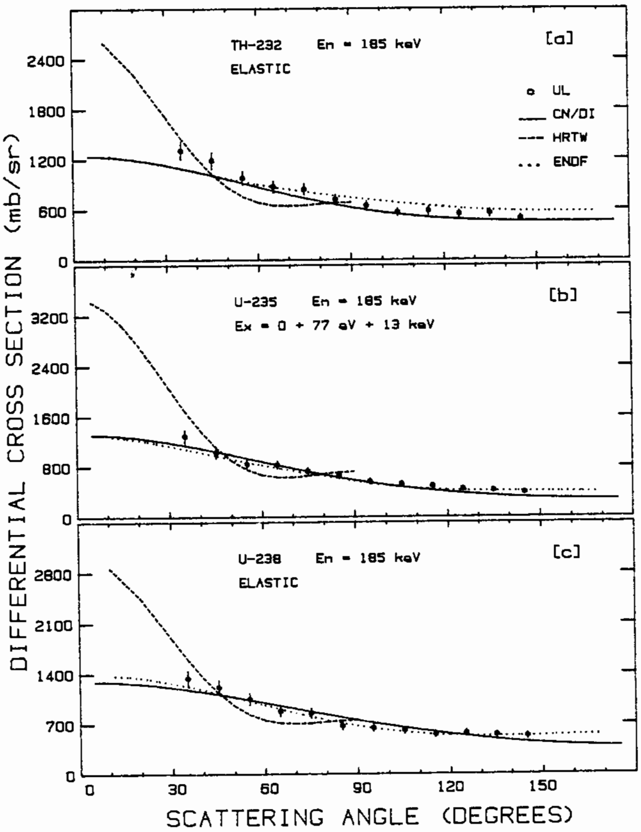


Fig. 4 Neutron elastic scattering angular distributions at E_n = 185 keV for (a) ^{232}Th, (b) composite ^{235}U, (c) ^{238}U, contrasting Lowell data[30] with theoretical analyses and ENDF/B-V evaluations.

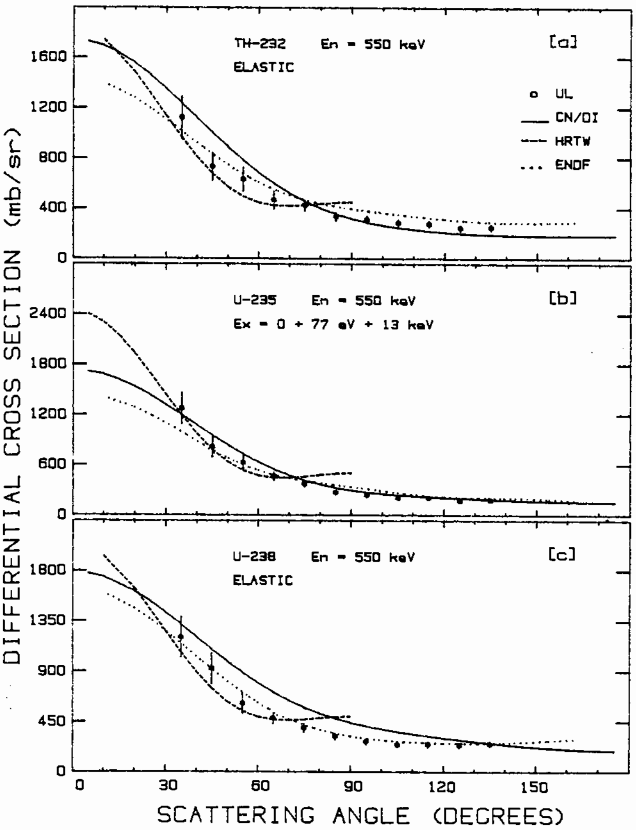


Fig. 5 Neutron elastic scattering angular distributions at E_n = 550 keV for (a) ^{232}Th, (b) composite ^{235}U, (c) ^{238}U, contrasting Lowell data[30] with theoretical analyses and ENDF/B-V evaluations.

Because the structure of elastic angular distributions becomes increasingly pronounced at higher incident energies (as is evident from, *e.g.*, the Bruyères results[17-27] from 0.6 to 3.4 MeV, a selection of which have been included in Figs. 6 - 8, and from previous Lowell findings,[15] incorporated within Fig. 6(b), for neutrons on ^{238}U at 2.5 MeV), and in view of the paucity of *inelastic* data to higher (vibrational) states in these nuclides, the main thrust of the Lowell studies has now proceeded to these higher energies.[37] To illustrate the current investigations, Fig. 6 *(at right)* shows results for ^{232}Th and ^{238}U(n,n) angular distributions at 2.4 and 2.8 MeV, while Figs. 7 and 8 *(below)* depict the progressive change in the structure (especially, peak-to-valley ratio) of the distributions for ^{242}Pu over the energy range $E_n = 0.6 - 3.4$ MeV.

It is clear from all the preceding that the standard (CN + DI) approach is able to render an admirable account of the variation of cross sections with energy and angle for neutron *elastic, as well as inelastic, scattering on the actinides.* However, the more fundamental HRTW formalism and calculations continue to warrant more detailed scrutiny and development, such as is now being pursued internationally, *e.g.*, at Lowell, Los Alamos (E.D. Arthur), Ohio (R. Finlay:OPSTAT), Oxford (P. Hodgson, M. Chadwick: WILMORE6) and Kiev (V.Plujko).

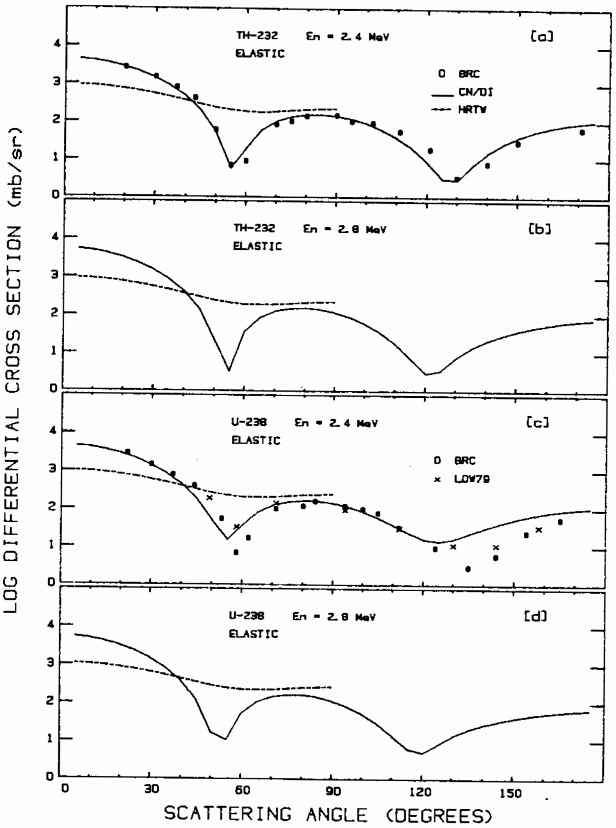
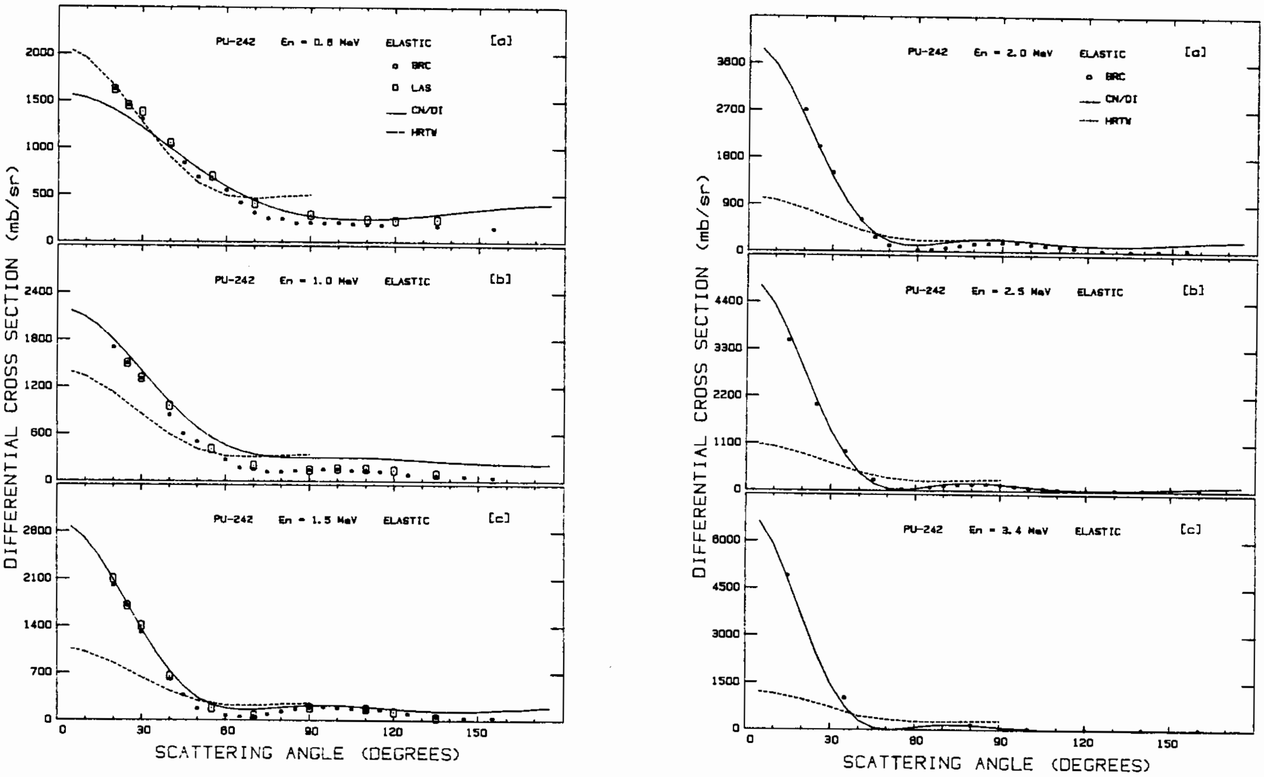


Fig. 6 *(right)* Neutron elastic scattering angular distributions at $E_n = 2.4$ & 2.8 MeV on (a,b) ^{232}Th and (c,d) ^{238}U, calculated in the standard (CN/DI) formalism *(solid curves)* and statistical S-matrix (HRTW) formalism *(dashed curves)*. The experimental data points depict measurements by the Lowell (LOW79)[15] and Bruyères (BRC)[17-27] groups (at 2.5 MeV), together with latest results[37].



Figs. 7 & 8 Progressive change with increasing incident energy from 0.6 to 3.4 MeV of the angular distributions for elastic neutron scattering on ^{242}Pu, contrasting the experimental data of the Bruyères (BRC)[27] and the Los Alamos (LAS)[36] groups with the predictions of standard (CN/DI) theory *(solid curves)* and statistical S-matrix (HRTW) theory *(dashed curves)*. The results graphically display the superiority of angular distributions over excitation functions as a sensitive test of formalism and mechanism: the angular integrations inherent in building excitation functions entail a $\sin\theta\, d\theta$ term which suppresses the contributions at forward angles ($\theta \leq 30°$), where the discrepancies between experiment and theory are most evident.

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TABLE I. Bruyères optical potential and deformation parameters, and Gilbert-Cameron level-density parameters.

Parameters	Nuclides					
Optical model	^{232}Th	^{235}U	^{238}U	^{240}Pu	^{242}Pu	^{244}Pu
Real potential V (MeV)	46.4 - 0.3E_n	46.4 - 0.3E_n	46.2 - 0.3E_n	46.14 - 0.3E_n	46.03 - 0.3E_n	45.92 - 0.3E_n
Imaginary potential W	3.6 + 0.4E_n	3.3 + 0.4E_n	3.6 + 0.4E_n	3.57 + 0.4E_n	3.51 + 0.4E_n	3.45 + 0.4E_n
Radius parameter r_0 (fm)	1.26	1.26	1.26	1.26	1.26	1.26
Diffuseness a (fm)	0.63	0.63	0.63	0.63	0.63	0.63
Radius parameter r_0' (fm)	1.26	1.26	1.26	1.26	1.26	1.26
Diffuseness a' (fm)	0.52	0.52	0.52	0.52	0.52	0.52
Spin-orbit pot. V_{so}	6.2 MeV	6.2 MeV	6.2 MeV	6.2 MeV	6.2 MeV	6.2 MeV
SO radius param. $(r_0)_{so}$	1.12 fm	1.12 fm	1.12 fm	1.12 fm	1.12 fm	1.12 fm
SO diffuseness a_{so}	0.47 fm	0.47 fm	0.47 fm	0.47 fm	0.47 fm	0.47 fm
Deformations						
Quadrupole β_2	0.190	0.220	0.198	0.200	0.204	0.242
Hexadecapole β_4	0.071	0.080	0.057	0.062	0.051	0.047
Level-density parameters						
$U_0 = Q(n,\gamma)$	4.956 MeV	6.467 MeV	4.784 MeV	5.412 MeV	5.047 MeV	4.590 MeV
Energy E_0	0.061 MeV	-0.563 MeV	-0.109 MeV	-0.170 MeV	-0.117 MeV	-0.224 MeV
Temperature T	0.387 MeV	0.397 MeV	0.392 MeV	0.407 MeV	0.400 MeV	0.399 MeV
Pairing energy P	1.35 MeV	0.69 MeV	1.15 MeV	1.04 MeV	1.11 MeV	1.00 MeV
Energy parameter a	29.44 MeV^{-1}	28.18 MeV^{-1}	28.71 MeV^{-1}	26.93 MeV^{-1}	27.78 MeV^{-1}	27.80 MeV^{-1}
Spin parameter σ	5.68	5.64	5.69	5.61	5.67	5.68
Shell correction S	0.75 MeV	-0.01 MeV	0.07 MeV	-0.85 MeV	-0.57 MeV	-0.66 MeV
Tangency energy E_x	4.50 MeV	3.83 MeV	4.28 MeV	4.17 MeV	4.23 MeV	4.11 MeV