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REVIEW OF (n;n',γ) CROSS-SECTION DATA ON ACTINIDE NUCLEI*

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ABSTRACT

The available high-resolution (n;n',γ) cross section data on actinide nuclei, essentially ^{238}U and ^{232}Th, are reviewed by dividing the measurement process into two steps. First, the measurement of discrete photon production cross sections is discussed by comparing white and monoenergetic neutron source techniques. Resultant cross sections are compared and their accuracies assessed. Second, the step of inferring inelastic scattering cross sections from photon production cross sections is discussed with particular emphasis on the problems connected with monopole transitions, low-energy transitions, prompt fission photons, and simple rotational models. The uncertainties introduced in this step are probably larger than those of the first.

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I. INTRODUCTION

Inelastic scattering on actinide nuclei provides one of the main moderation processes for fast neutrons in reactors; consequently, accurate knowledge of these cross sections (CS) are required, particularly those of ^{238}U. Direct detection of scattered neutrons provides the best source of such needed inelastic data since the process of interest is straightforwardly measured. However, the intensities and time resolutions of the monoenergetic, pulsed-neutron, time-of-flight (TOF) facilities presently providing these CS are limited so that inelastic groups from closely-spaced levels cannot be resolved. The development of high-resolution semiconductor photon detectors has allowed the extension of inelastic scattering studies to closely-spaced levels through the measurement of production CS of de-excitation photons. The resolution of these measurements is only limited by the photon-detector-resolution, ~ 2 keV; however, this method is indirect and extraction of accurate inelastic-scattering CS from such data requires detailed and accurate spectral information on the various decays into and out of the levels of interest.

This paper reviews the present state of these $(n;n',\gamma)$ measurements of discrete photon groups in actinide nuclei. The various overall photon production measurements with low-resolution detectors are not considered. Some aspects of actinide $(n;n',\gamma)$ measurements have been discussed in recent review papers.[1-3] This paper will emphasize the special difficulties associated with actinide $(n;n',\gamma)$ measurements and their interpretation. Many of these difficulties do not exist for lighter targets.

The $(n;n',\gamma)$ data on actinide nuclei are very limited. Much of this data dearth results from the inherent difficulty of measuring CS for weak $(n;n',\gamma)$ photon groups in the presence of photon and neutron fluxes from open fission channels in the nuclei under study. No Ge(Li) $(n;n',\gamma)$ measurements appear in the CINDA index for $^{233,235}U$ or $^{239,240,242}Pu$. Four CS measurements have been reported for ^{238}U: Van de Graaff results from Lowell[4] and SUNI,[5] and white source results from ORELA[6] and KFK.[7] Lowell[8] and SUNI[9] have also reported results for ^{232}Th.

Production CS for photons from the excited members of the ground-state, (GS) rotational bands of ^{238}U and ^{232}Th have not been measured. These CS for the 2^+ and 4^+ levels of ^{238}U are the most important actinide inelastic data for reactor application, particularly below $\sim$ 0.5 MeV which is difficult for the existing monoenergetic TOF facilities. The 2^+ and 4^+ deexcitation photons of ^{238}U have internal conversion coefficients of 620 and 12, and target attenuation half-distances of 0.05 and 0.5mm, respectively, which makes them exceedingly difficult to observe experimentally. This difficulty with low-energy photon detection also exists of course for low-energy transitions between higher excited states.

The problem of determining inelastic scattering CS from actinide $(n;n',\gamma)$ measurements divides into two somewhat separate steps. The next section of this report discusses the problems of the first step which is to accurately measure photon production CS. Section III discusses the problems of the second step which is to accurately determine inelastic scattering CS from the photon CS. The last section of this report summarizes the conclusions.

II. MEASUREMENT OF PHOTON PRODUCTION CS

A. Experimental Apparatus and Procedures

Most actinide (n;n',γ) measurements have been made at 125°. The angular distributions $\sigma_\gamma(\theta)$ of the de-excitation photons are an expansion of even-order Legendre polynomials; that is, $\sigma_\gamma(\theta)=a_0+a_2p_2+a_4p_4+\ldots$ where the maximum order is twice the decaying-level spin. For most decays the p_4 and higher terms are thought to be negligible; consequently, the angle integrated photon CS are simply 4π times the measured differential CS at 55° or 125°. Egan *et al.*[8] have calculated proton angular distributions for ^{232}Th with statistical model calculations. Their results indicate that this neglect of the p_4 and higher terms introduces at worst a 1% error for levels above the GS band.

The experimental arrangement used for the Lowell ^{232}Th and ^{238}U measurements,[4,8] is shown in Fig. 1 and is more or less standard for monoenergetic-source measurements. A pulsed proton beam from a 5.5-MV Van de Graaff impinged on a wobbled-and-water-cooled solid tritium target producing nearly monoenergetic neutrons. A 3.8-cm diameter and 1.2-cm thick ^{238}U disc was located at 8.5 cm from the neutron source and deexcitation photons were detected by a 40-cm^3 Ge(Li) detector placed at 125° and 88 cm from the center of the scatterer. A NaI annulus surrounded the Ge(Li) detector, and was used to detect Compton events. Shielding of these two detectors was accomplished using structures of lead, a lithium carbonate and paraffin mixture and a copper shadow bar. A paraffin block was placed between the scatterer and the Ge(Li) detector to reduce the neutron flux on the detector. Neutron monitoring during the data runs was done by a plastic scintillator. The neutron fluence was measured after each run with a long counter and a fission chamber in a TOF system. The absolute efficiencies of these detectors were determined by comparison to a recoil-proton telescope whose absolute efficiency was known.

A typical Ge(Li) time spectrum is illustrated in Fig. 2 which indicates the reduction of the neutron events due to the paraffin plug. These neutrons are separated by 47 ns from the photons. Two 4096-channel energy spectra were accumulated simultaneously in a multichannel analyzer. One was the time-gated, energy-gated and Compton-suppressed spectrum yielding the accepted data. The other spectrum contained those photon events which fell in the time window but were rejected by the annulus. Figure 3 is a simplified representation of a ^{232}Th Compton-suppressed photon spectrum and the spectrum of events rejected by the Compton-suppression system but accepted in the time window in the Ge(Li) TOF system. Lines marked (b) are due to room background and to natural radioactivity in the thorium sample. Unmarked lines are from the (n;n',γ) reaction. The lower spectrum is plotted on the same scale and was obtained during the same run as the upper one. With this apparatus the Compton background is reduced by a factor of four; however, the NaI annulus is very sensitive to background radiation and must be heavily shielded to avoid large deadtime effects. None of the Th(n;n',γ) lines were observed in the rejected spectrum.

White source measurements are essentially similar; however, photon spectra for all incident neutron energies are measured simultaneously. Figure 4 shows the apparatus used at ORELA for ^{238}U.[6] Eight hundred 35-nsec FWHM neutron pps from ORELA were filtered through ^{10}B to inhibit time overlap and 14.0 cm of Th to attenuate gamma flash. The flux was collimated into a $\sim$ 7-cm diam beam which traversed a proton-recoil radiator foil at 21.430 m and a ^{238}U scattering sample at 22.181 m consisting of a 11.11-cm diam, 0.76-mm thick, disc. Photons were counted at 125° with a 95-cm^3 Ge(Li) detector with its front face 28 cm from the sample center and face-shielded with 1.8 mm of lead. Simultaneously the neutron flux was measured with a recoil-proton telescope described in detail in Ref. 10. Fast timing pulses were obtained from the Ge(Li) detector with a constant fraction discriminator operated with slow rise-time reject and from the proton-recoil

detector with a crossover timing discriminator. From these fast pulses neutron TOF for both detectors were measured with an EG&G TDC100 time digitizer operated in the single stop mode. Simultaneous slow pulses produced 2048- and 256-channel pulse-height spectra, respectively, for each of 160 timing bins.

Both ^{238}U and ^{7}Li photon production CS were measured by alternating the required samples in and out of the ORELA beam with a period of $\sim$ 1 day. Figure 5 shows a Ge(Li) pulsed height summed, TOF spectrum from the ^{7}Li target. The γ-flash photon group occurs at a clocktime of 1.3 μs and has a FWHM of 35 ns. Photons of interest produced by 5.0 to 0.5 MeV neutrons span clocktimes from 1.9 to 3.5 μs, respectively. This spectrum also contains neutrons. In particular the peak at 4.4 μs is from neutron scattering from the 0.25-MeV ^{7}Li resonance. Neutrons can be seen in the Ge(Li) pulse-height spectra. Figure 6 shows a ^{238}U spectrum produced from 1.21-to-1.49-MeV neutrons. The broad peaks are attributed to Doppler broadened deexcitation photons produced from neutron inelastic scattering from the Ge of the detector.

B. Uncertainties in Photon-Production CS

From 125° spectra, 4π photon production CS at an incident neutron energy E_n for discrete photons of energy E_γ can be calculated from the expression,

$$\sigma(E_\gamma,E_n) = N(E_\gamma,E_n)m(E_\gamma,E_n)/nF(E_n)\varepsilon(E_\gamma) \qquad (1)$$

where $N(E_\gamma,E_n)$ is the deadtime-and-pileup-corrected photo-peak yield, $m(E_\gamma,E_n)$ is the finite-sample-size correction, $F(E_n)$ is the neutron fluence $\varepsilon(E_\gamma)$ is the photon detection efficiency with respect to 4π steradians, and n is the sample thickness. All the terms of Eq. (1) contribute to the uncertainty in $\sigma(E_\gamma,E_n)$ although that of n should be negligible. The dependence of these terms on E_γ and E_n makes, in principle, the uncertainty analysis of Eq. (1) rather complicated. For example, ε is fully correlated over E_n for a given E_γ, whereas F is fully correlated over E_γ for a given E_n. Most authors, however, have simply estimated an overall normal-

ization error fully correlated over both E_n and E_γ and an uncorrelated error for each datum.

For isolated intense lines under ideal conditions the full-energy peak yields should be obtainable to ∿ 2% accuracy excluding the counting statistical contribution. However, the complicated spectra obtained from neutron bombardment of radioactive actinide targets are far from ideal. In addition, counting losses for deadtime, pileup etc. must be accurately accounted for. The single largest contribution to the uncertainty in the ORELA ^{238}U CS is from pileup and area extraction from asymmetric line shapes.[6] In particular, the counting rate and line shape from the Ge(Li) detector were monitored with 662-keV photons from a ^{137}Cs source near the sample. After the clock deadtime correction, events from this source showed no counting losses at long flight times; however, at the short flight times of interest peak tailing and a consistent 10% counting loss from pileup and electronic deadtimes were measured. A 5% error was assigned to the peak yields to cover these effects.

The second large source of systematic uncertainty is the fluence determination or equivalent normalization. The fluence used in the Lowell measurements[4,8] was monitored with a long counter and fission chamber which were ultimately normalized to a recoil proton telescope whose absolute efficiency was known. The neutron flux for the Karlsruhe measurement[7] was determined with a NeI02 plastic scintillator. The proton recoil telescope used at ORELA is capable of determining the fluence to 3 to 4% accuracy. Measurements could also be made relative to a standard photon-production CS such as ^{56}Fe(847 keV), or ^{7}Li(478 keV). These reference CS are known to the order of 8% uncertainty. The ^{7}Li 478-keV photon is emitted isotropically but is significantly Doppler broadened. The ^{56}Fe 847-keV CS has a significant p_4 term in its angular distribution and contains measureable resonance structure.[11] The SUNI data are normalized to the ^{56}Fe photon CS. The flux could also be measured with a U^{235} fission chamber; in the 1-to-2-MeV range the ^{235}U fission CS is thought known to ±5%.

The absolute efficiency of the GeLi detector system should be measurable with standard sources as a function of photon energy to 2-4% uncertainty. Care must be taken to insure that the calibration line shapes are similar to those obtained in the actual measurement. The efficiency calibration must, of course, incorporate threshold effects from discriminators used to obtain timing pulses. The finite sample corrections for neutron flux attenuation, photon attenuation, finite geometry and multiple scattering are discussed in Ref. 1-3. For measurements on actinide nuclei at neutron energies above the fission threshold the inelastic photons from fission neutrons must also be considered. For reasonably energetic photons and reasonably small samples the uncertainties from finite-sample-size corrections should be less than 2%.

Under ideal conditions and with careful attention to details $\sigma_\gamma(E_n,E_\gamma)$ could be measured on actinide nuclei with a systematic uncertainty of ±5%. To this, of course, should be quadratically added the statistical uncertainty which for intense lines should be small. Typical $(n;n',\gamma)$ CS on lighter nuclei are generally accurate to ±10%, as determined from comparisons to directly measured inelastic CS.[1-3] Present $(n;n',\gamma)$ measurements on ^{238}U and ^{232}Th have quoted total uncertainties on the order of 10%.

C. Photon Production Cross Sections

Figure 7 shows ^{232}Th photon angular distributions measured at SUNI[9] using a ^{7}Li(p,n) source. According to the authors[9] the analysis of photon-angular distributions can be useful, in some cases, for spin assignments but in general the admixture of multipoles does not allow unique fits to the measured angular distribution shapes. The observed data have to be corrected for a skewing of the angular distributions which results from neutron and gamma attenuation in the sample. The skewing correction factor is given on the upper right of Fig. 7 and was calculated from the measured data at θ=45° and 135°. The measured distributions corrected in this manner fit the computed angular distributions for the assigned spin values.

The ^{232}Th measurements at 90° from SUNI[9] are compared with those of Lowell[8] in Fig. 8. The solid lines approximate the (n;n',γ) data of SUNI and should generally be increased or decreased by 0 to 15% if the photon angular distributions were properly included.[9] The data points with error bars are the 125° results of Lowell and the dashed curves are model calculations. Figure 8 shows inelastic scattering CS; however, both groups used essentially the same decay schemes so the results on this basis should be comparable. However, the fact that the SUNI CS were derived from 90° measurements confuses any detailed comparison.

A more definitive comparison can be made for ^{238}U. Photon production CS from the four 125° measurements for eight transitions are shown in Figs. 9 and 10, where the plotted errors are uncorrelated uncertainties. The triangular data points are the results from Lowell.[4] They estimate an additional 7% normalization uncertainty for their CS. No datum has an uncorrelated uncertainty less than 6% so each datum has an absolute uncertainty greater than $\sim$ 9%. Similarly, the ORELA CS shown as circles are assigned a 6% normalization uncertainty.[6] A 5% minimum uncorrelated uncertainty gives at least an 8% uncertainty for each datum. The KFK CS shown as squares have an additional 11% normalization uncertainty[7] and the SUNI CS shown as x's without uncorrelated uncertainties have a 10% normalization uncertainty.[5]

Clearly, the KFK data seem discrepantly low with respect to the other three measurements for the strong transitions of Fig. 9. In particular, the sum of the 635, 584, 1015, 1061, 886, and 952 CS from KFK in the 1.2-to-1.5-MeV region are $\sim$ 35% smaller than the corresponding summed CS from the Lowell data. The corresponding sums for the ORELA and SUNI data are equal within a few percent and are $\sim$ 10% smaller than the Lowell summed CS. The systematic difference between the CS depends upon the transition. These differences may be caused by peak-area extraction procedures and confusion with other unresolved photon groups. In general the Lowell CS are from 0 to 20% larger than those of ORELA, whereas the SUNI CS are both higher and lower than the ORELA CS depending on the transition. For example, the ORELA CS for the 635-keV photon line are $\sim$ 20% smaller than those of SUNI, whereas for the 1015 keV photon line the opposite is true. A suitable average of the Lowell, SUNI, and ORELA photon-production CS on a transition by transition basis would perhaps provide CS accurate to ±10%, at least for the strong transitions.

D. Comparison of White- and Monoenergetic-Source Methods

Measurements have been made with both monoenergetic and white neutron sources. Each method has its own particular advantages and disadvantages. The Lowell[4,8] and ORELA[6] ^{238}U measurements required similar data acquistion times; however, the ORELA photon spectra were all measured simultaneously and normalized together, whereas the Lowell spectra were measured one at a time, each with its separate normalization. This difference is reflected in the smoothness of the CS shown on Figs. 9 and 10. This smoothness does not, however, necessarily imply a more accurate normalization. The monoenergetic sources can focus all the neutrons into narrow energy band which facilities setting up and is a real advantage for a concentrated effort to study weak transitions at a single neutron energy. On the other hand, for an excitation function of the more intense lines a white source could be superior. Timing resolution is not crucial for either method.

Ignoring shielding, the background around the detector in a white source measurement should be substantially less than that from a monoenergetic source. However, this advantage is more than compensated for by the facility to discriminate against off-time background events with monoenergetic sources, particular neutrons scattered directly into the detector. For example, the broad peaks in Fig. 6 from Ge inelastic scattering do not appear in Fig. 3 because of time discrimination. However, the ORELA ^{238}U measurement was not optimized: in a recent $^{56}Fe(n,n'\gamma)$ measurement these broad peaks were reduced by a factor of five with a LiH plug and massive shielding.[12] Nevertheless this time discrimination is an important consideration.

The "in-beam" count rate of white source measurements can be substantially larger than those from monoenergetic sources. Ignoring backgrounds, if one assumes that a 12 hr Lowell run at 5MHz with a 30 ns time gate gives an equivalent spectrum as a 36-ns time bin from the 20 day ORELA run at 800 pps,

then the ORELA "in-beam" spectral count rate is ten-fold faster than that of Lowell. In addition, the ORELA spectra were measured concurrent in time. For this ORELA run the flux was attenuated by a factor of $\sim$ 20 to reduce the γ-flash and count rate to a tolerable level. This has some consequences. Even ignoring the γ-flash problem ORELA data has substantially larger dead-time and pileup corrections problems than the Lowell data. However, for the same reason target activation has a smaller effect on the spectra.

White source systems have other advantages. The spectra are normalized and taken together. In addition the stability and large geometries of white sources provide the potential for superior energy definition and normalizations. Moreover, relatively thin samples with large areas can be employed allowing significant simplifications of finite-sample-size corrections. Finally white sources have the ability to measure photon spectra at low incident neutron energies. To date neither method has been able to convencingly measure thresholds of weak transitions or production CS for low-energy transitions.

III. CALCULATION OF INELASTIC CS FROM PHOTON CS

Three of the four $^{238}U(n;n',\gamma)$ measurements give photon CS which agree within the ±15% level of uncertainty for intense lines. Nevertheless published inelastic CS from these measurements show considerable discrepancies. These discrepancies result largely from different assumptions concerning the ^{238}U level-decay scheme. In principle, the calculation of inelastic CS from photon CS requires a complete knowledge of the decay properties of all levels populated either directly or indirectly by the incident neutrons. In particular, accurate branching ratios, BR; EO decay intensities; and multipolarity mixing ratios are required. The uncertainties and confusion in level decay information may contribute larger errors to the desired inelastic CS than those of the measured photon CS. The errors on published inelastic CS contain no contribution from these spectral uncertainties. In addition, the problem of feeding from higher levels are largely ignored. This section discusses some of these problems using almost exclusively those of ^{238}U as an example. The decay scheme problems for other actinide nuclei would probably be similar.

A. The ^{238}U Level-Decay Scheme

Figure 11 shows part of the adopted ^{238}U energy level scheme from the 1977 Nuclear Data Sheets compilation of Ellis.[13] All of the nuclear structure information on ^{238}U were collected, compared and evaluated resulting in the Fig. 11 proposed level diagram and band structure. This should be the "best" level scheme for ^{238}U. All levels up to 1200 keV of excitation with spins ≤ 5 are shown except for a possible 2^+ state at 998.3 keV. The available $^{238}U(n;n',\gamma)$ data does not contridict this level structure.

Unfortunately the Nuclear Data Sheets compilations do not result in an adopted decay scheme with BR; however, most of the available information on BR is listed in Ref. 6 and 13. In addition to the $(n;n',\gamma)$ CS results there are the photon branching ratios from $(n;n',\gamma)$ spectrum of Demidov *et al.*[14]

the ^{238}Pa β^- decay study of Trautmann *et al.*[19] and the Coulomb excitation work of McGowen *et al.*[16] Most inelastic ^{238}U CS were calculated without attempting to incorporate decay scheme information from nuclear models and other types of spectral measurements. The ORELA inelastic CS contains, to some extent, other ^{238}U spectral information.

Comparison of BR from different measurements shows discrepancies and conflicts even for high-energy transitions. These BR should be energy and measurement independent. Table I list 449-keV to 1083-keV BR from the 1128.7 keV (2^-) level from varous measurements. Most of these should be accurate to ±10%. Nevertheless, the spread is considerable with an average value of 1.17 and an internal uncertainty of ±19% which is not reflected in inelastic CS errors. Some of the ^{238}U photon decays are known doublets. In particular, the 1059.5 and 1060.3-keV levels as well as the 1167.7-1169.1 keV levels are nearly degenerate in energy and the 997.5 keV 3-level may be doublet with a 2^+ state.

Table I. BR of the 449 keV to 1083 keV BR From the 1128.7 keV (2^-) Level

Measurement	Ref.	BR
Lowell	4	1.20
SUNI	5	0.89
Demodiv	14	0.41
ORELA	6	0.90
β^-	15	1.36
Coulomb	16	1.26
Average		1.17+0.21

B. Low-Energy Branches

The low-energy transitions are particularly troublesome for calculating U^{238} inelastic CS. The BR and even existance of these transitions are uncertain. Moreover, the unknown multipolarities of some of these transitions provide additional uncertainty through their interval conversion coefficients. Most of the information on these transitions comes from the single ^{238}Pa β^- Ge(Li) spectrum of Trautman *et al.*[15] In general, most of these branches have not been clearly seen in (n;n',γ) measurements, partly perhaps because most of these measurements have been optimized for higher-energy photons.

Figure 12 illustrates some of the problems and shows possible inelastic CS to the (2^-) level at 950 keV. The only high-energy transition from this level is the 905-keV photon to the GS band 2^+ level. Assuming only thid decay and no feeding the resultant Lowell (triangles), SUNI (x's) and ORELA (circles without error bars) CS are shown in Fig. 12. However, the β^- decay work indicates possible 270 and 218-keV decays from this level to the octupole 1^- and 3^- levels and one decay into this (2^-) level from the higher 1128.7 keV ($2^{-\prime}$ level. Assuming these transitions exist the inelastic CS from ORELA data are shown in Fig. 12 on the lowest CS circular data points with error bars.

These transitions increase the inelastic CS by $\sim$ 40%. However this CS assumes that the ORELA estimate of $\sim$ 0.20 for the 270 and 218-keV BR were correct. Trautman *et al.*[15] measured these BR to be $\sim$ 0.5, whereas Domidov *et al.*[16] measured these BR to be $\sim$ 1.0. Use of the Trautman *et al.* BR would increase the inelastic CS by another 50%. Moreover, these low-energy transitions can be mixed E2/M1 and their mixing ratios are unknown. For the ORELA inelastic CS they were all assumed to be E2. If the low-energy branches were all assumed to be M1 the interval conversion coefficients would increase by about a factor of five and the 950 keV inelastic CS would increase by about a factor of two

to the largest inelastic CS shown in Fig. 12. The error in the 950-keV inelastic CS from spectral uncertainty is a factor of $\sim$ 3.5! Unfortunately the corresponding transitions do exist for the exhaustively studied ^{234}U decay scheme and are mixed E2/M1.

Inclusion of these low energy transitions, however, simply transfer inelastic CS from one level to another nearby level. For example, inclusion of the 218-keV decay, assumed to be E2 with a 0.20 BR, transfers 0.023b of inelastic CS in the 1.5-to-2.0-meV region from the 732-keV level to the 950-keV level. The sum of the inelastic CS to these two levels is the same with or without this branch. In fact, the sum of the photon CS to the GS band, corrected for E0 transitions, is the sum of the inelastic CS to these higher excited states. Figure 13 compares such a sum for ^{238}U with the ENDF/B-V evaluation[17] for all levels above the GS band and below 1.2 MeV of excitation. For 1.2 MeV neutrons, above which their are no feeding corrections, the summed measured cross section is in excellent agreement with the ENDF/B-V evaluated value, whereas at the ENDF/B-V maximum value of 1.75 barns the sum is approximately 10% low, about the limit of the experimental accuracy. Inelastic CS can escape undetected from the higher-energy levels through E0 transitions.

C. Monopole Transitions

Monopole transitions do exist in heavy nuclei and may compete with higher-order multipole de-excitation. These transitions between levels of the same spin occur through wave-function components with the same K and in the simple Bohr-Mottelson rotational model exist only for transitions from the β-band to the GS band. The information available on E0 transitions prior to 1972 are collected, discussed, and compared with theory in Ref. 18. More detailed and recent information for actinide nuclei can be obtained from *Nuclear Data Sheets* for ^{209}At, ^{212,214}Po, ^{228,230,232}Th, and ^{232,234,238}U. In particular, McGowen *et al.*[16] have deduced that over 50% of the excitations of the ^{238}U β-band 2^+ level at 1037 keV decay by E0 radiation and that E0 decay from the ^{232}Th β-band 2^+ level at 774 keV is 16.7 times more intense than the corresponding E2 photon decay.

Figure 14 shows inelastic CS measured at SUNI to the 0^+, 2^+, and 4^+ rotational members of the ^{232}Th β-band.[9] The open circles with error bars are CS from direct measurement, whereas the points without error bars are the corresponding CS deduced from (n;n',γ) measurements. The solid curve is a statistical model calculation. The authors attribute much of the CS discrepancy between the two methods to E0 transitions which is extremely plausible; particularly since McGowen *et al.*[16] have measured an E0 decay from this 2^+ level. Unfortunately this 2^+ level cannot be resolved from a 3^- level.

Figure 15 shows inelastic CS from ^{238}U(n;n',γ) measurements for the β-band, 2^+ level at 1037 keV. The three E2 decays from this level to the GS band are observed in (n;n',γ) spectra and the Lowell[4] inelastic CS (triangles) is simply the sum of the photon CS for these branches. The corresponding sum for the ORELA data[6] is shown as the circles without error bars. The circles with error bars are the ORELA inelastic CS which includes the E0 BR of 4.4±1.2 as measured by McGowen *et al.*[15] This BR increases

the inelastic CS by over a factor of two and its uncertainty introduces a 15% fully-correlated uncertainty in the CS. This uncertainty dominates those from the photon CS. In addition, Ellis[13] speculates that perhaps 67.5% of the excitations of the 966-keV 2^+ level decay by the E0 mode. This decay is not included in the Fig. 13 measured CS and would contribute 0.5b of inelastic CS at 1.5 MeV! Moreover, no photon decays are observed from the β-band 0^+ and 4^+ levels, perhaps because they decay mostly by E0. About 70% of the excitations of the ^{234}U β-band 0^+ level decay by the E0 mode. Clearly, the lack of detailed knowledge on E0 transitions is a major source of uncertainty for inelastic CS deduced from actinide $(n;n',\gamma)$ measurements.

D. "Prompt" Photon From Fission

"Prompt" photon from fission can provide a significant source of uncertainty and confusion at neutron energies above the fission threshold. Some aspects of this problem are illustrated in Fig. 16 which shows pulse-height spectra for 1.0-to-1.4-MeV photons as a function of neutron energy from a white-source ^{238}U measurement.[6] The neutron energy range and corresponding flight-time intervals are given on the figure. Verbinski *et al.*[19] have measured with NaI that thermal fission of both ^{235}U and ^{239}Pu give about 2 prompt (< 10 ns) photons per fission per MeV in the 1.0-to-1.5-MeV range. Assuming that fast-fission of ^{238}U does the same, the full-energy photopeak area from prompt fission photons can be estimated and is shown as the hatched area on the bottom spectrum of Fig. 16. For this spectrum about 300 full energy events per channel, on average, may be from prompt fission photons. These and the corresponding Compton events provide a significant background.

More importantly these protons can appear in discrete lines, Kahn *et al.*[20] have measured Ge(Li) spectra of prompt (< 2 ns) photons from thermal ^{235}U fission and Sand *et al.*[21] have measured Ge(Li) spectra from 20 ns to 1 μs after thermal fission of both ^{235}U and ^{239}Pu. Some weak lines very-weak lines which appear on $^{238}U(n;n',\gamma)$ spectra above the fission threshold have energies corresponding to those measured from prompt fission. In particular, the 1279.8 keV ^{134}Te 2^+ decay, shown as a shaded area in Fig. 16, is seen in about 2% of the thermal fissions of ^{235}U and ^{239}Pu with a half life of ∿ 160 ns. Much or most of the area of this line in the 2.0-to-4.3-MeV spectra could be accounted for as a fission photon. Moreover, because of its 160 ns half life, this line appears in the upper spectra of Fig. 16 where its production as an inelastic scattering photon is energetically forbidden. The other shaded

line at 1180.8 keV corresponds to another well known, but weaker, fission isomer (∿ 600 ns) and also appears at forbidden neutron energies. Unfortunately both of these lines are doublets with de-excitation photons from levels excited in inelastic scattering.

These full-energy peak areas result from a first-plateau CS of ∿ 0.5b above a ∿ 1.5-MeV threshold. The problem is less severe for ^{232}Th which has about the same fission threshold but only ∿ 0.1b first-plateau CS. The ∿ 0.5 MeV fission thresholds and ∿ 1.5b first-plateau CS of ^{240}Pu and ^{242}Pu could make (n;n',γ) measurements using present techniques difficult for these isotopes except for intense lines. The fissile isotopes of ^{233,235}U and ^{239,241}Pu also have 1-to-2b fission CS in the 0.1-to-1.0-MeV region. In addition, the fission-photon background from thermalized background neutrons could be severe. Perhaps if the photon detectors were anticoincidenced with fission-chamber targets this fission-photon background could be alieviate, particularly for fissile samples. This would require working with samples of only a few grams.

E. The ^{238}U Octupole Band and Rotational Models

Figure 17 shows the magnitude of the feeding corrections from Ref. 6 for members of the K=0 octupole band of ^{238}U. The 1^-, 3^-, and 5^- members are fed by 7, 6, and 1 branches, respectively, and the intraband E2 decays where calculated assuming a simple rotational model with the GS band intrinsic quadrupole moment. Moreover, the $5^- \rightarrow 4^+$ and $1^- \rightarrow 0^+$ crossover transitions to the ground state band are nearly degenerate in energy, and the doublet intensity was divided using Alaga's rule;[22] which relates crossover transition intensities between good rotational bands. That is,

$$\frac{B(L;II \rightarrow IF)}{B(L;II \rightarrow IF')} = \left[\frac{\langle II,L,KI,KF-KI | IF',KF\rangle}{\langle II,L,KI,FK-KI | IF,KF\rangle}\right]^2 \qquad (2)$$

where the notation may be obvious and discribed in Ref. 23. Both bands must have K as a good quantum number.

In particular, below 1.5 MeV $\sigma_\gamma(3^- \rightarrow 4^+)/\sigma_\gamma(3^- \rightarrow 2^+) = 0.78 \pm .02$ which agrees with Alaga's rule of 0.81. Likewise, below the 5^- threshold at $\sim$ 1.0 MeV, $\sigma_\gamma(1^- \rightarrow 0^+)/\sigma_\gamma(1^- \rightarrow 2^+) = 0.62 \pm .02$ which also agrees with Alaga's rule of 0.61. Consequently Alaga's rule was assumed to be valid also for the 5^- crossover transitions between these bands and the separation of the doublet intensity into $5^- \rightarrow 4^+$ and 1^- to 0^+ components followed directly. Moreover, doublet intensity satisfyingly could simultaneously satisfy both branching ratios requirements as a function of neutron energy. Unfortunately the other excited bands of ^{238}U appear not to have pure K quantum numbers; that is, are mixed as probably should be expected, since in general the cross-over transitions do not follow Eq. (2). The smooth curve of up to 2 MeV on Fig. 16 are results of a statistical model calculation of Jary *et al.*[24] and fit the "measured" CS remarkably well. Figure 16 also illustrates another observation which is sometimes useful. Namely, that the cross section rise above threshold tends to be much sharper for low-spin states than for high spin states.

IV. SUMMARY AND CONCLUSIONS

To date, four ^{238}U and two ^{232}Th, neutron-energy-dependent, $(n;n',\gamma)$ measurements with Ge(Li) detectors have been reported. Similar results for other actinide nuclei have been published. Three of the four ^{238}U measurements give photon production CS which agree at the ±15% level, near their quoted uncertainties, for intense lines. For weak lines the agreement is less good. The ^{232}Th data are shown in Fig. 8. A suitable average of both the ^{238}U and ^{232}Th data would perhaps provide more accurate photon CS. With careful attention to detail 5% measurements should be possible.

Measurements have been made with both monoenergetic and white neutron sources. Each type of system has its own particular advantages. A monoenergetic system by definition produces all the neutrons in a narrow energy bin allowing easy set-up and important discrimination against off-time events. In addition, the counting is essentially continuous in time. White-source systems are very stable and use large geometries allowing excellent neutron energy definition and the potential for superior normalizations. In addition, the real background should be smaller and thinner samples can be employed reducing the problems of finite sample corrections.

The main limitation at the present time in determining elastic CS from $(n;n',\gamma)$ measurements, at least for ^{238}U, is the lack of information on the decay modes which are not readily observable in $(n;n',\gamma)$ results. This information is required to accurately calculate inelastic CS from photon CS and part of the discrepancy between published inelastic CS is due to the large uncertainty in this information. In particular, E0 transitions intensities must be either measured or estimated with plausable uncertainties from systematically studies of the existing E0 data. The low-energy transitions, most of which for ^{238}U have only been observed in one ^{238}Pa β^- decay spectrum,

must be clarified: either from (n;n',γ) measurements tailered specifically for this purpose or other types of spectral measurements. In principle, the deduced inelastic CS are only valid up to the neutron energies at which the feeding corrections are understood. Above 1.2 MeV the neutron inelastic strength for ^{238}U seems to be shared among many weak levels whose decay are difficult to measure and easily confused with fission photons. For ^{233,235}U and ^{239,240,242}Pu this problem could be severe. Nevertheless, attempts should be made to determine these CS from (n;n',γ) measurements. The spectral problems for these actinides will probably be similar to those of ^{238}U which have been discussed. The feeding from higher levels is a severe problem. Perhaps some estimates could be made from statistical model calculations.

Considerable progress has been made in determining inelastic scattering CS for the levels above the GS rotational bands in actinide nuclei. A complete understanding of these CS with small uncertainties will require both direct scattering measurements, (n;n',γ) and auxillary spectral measurements, and interpolation and extrapolation with sophisticated model calculations.

ACKNOWLEDGMENTS

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FIGURE CAPTIONS

1. Experimental arrangement used for the Lowell ^{238}U and ^{232}Th measurements with a Van de Graaff p+t source.
2. Ge(Li) time spectrum from the Lowell measurements. The neutrons have been attenuated with a paraffin plug and are separated by $\sim$ 50 ns from the photons.
3. Simplified representation of a $^{232}Th(n;n',\gamma)$ photon spectrum measured at Lowell. The top spectrum is Compton suppressed; whereas the bottom spectrum are the events rejected by the Compton suppression system, without which the measured spectrum would have been the sum.
4. Experimental apparatus used for the ORELA ^{238}U measurement. The recoil-proton-detector radiator and scattering sample were 21.43° and 21.181 m, respectively, from the neutron-producing target.
5. Ge(Li) TOF spectrum from the ORELA $(n;n',\gamma)$ measurements on a ^{7}Li target which was alternated with a ^{238}U target. Photons of interest produced by 5.0 to 0.5-MeV neutrons span clock times from 1.9 to 3.5 μsec, respectively. Gamma-flash photons occur at 1.3 μsec and neutron scattering from the 0.25-MeV ^{7}Li resonance produces the peak observed at 4.4 μsec.
6. A ^{238}U $(n;n',\gamma)$ photon spectrum produced by 1.00-to-1.21-MeV neutrons from ORELA. The very broad groups are Doppler broadened photons produced by inelastic scattering from the Ge of the detector.
7. Comparison between the SUNI measured and calculated ^{232}Th $(n;n',\gamma)$ angular distributions.
8. Inelastic scattering CS of ^{232}Th measured at 125° by Lowell. The solid line approximates the corresponding 90° CS of SUNI. For these levels the two groups use essentially the same decay schemes. The dashed curves are statistical model calculations.

9-10. Comparison between measured 4π photon production CS from various ^{238}U (n;n',γ) measurements: KFK (squares), SUNI (x's without error bars), Lowell (triangles) and ORELA (circles). The error bars do not include normalization uncertainties.

11. Proposed ^{238}U rotational-band structure of *Nuclear Data Sheets*.[13] All levels below 1200 keV with spins less than five are shown except for a possible 2^+ state at 988.3 keV.

12. Possible inelastic CS to the proposed 2^- level at 950 keV. The Lowell (triangles) and SUNI (x's) CS are simply the photon CS for the 905-keV decay to the GS-band 2^+ level. The ORELA CS with this assumption are the circular data points without error bars. The circular data points with error bars are CS calculated from the ORELA data incorporating low-energy transitions. If these transitions are M1 the largest CS results and if they are E2 the lower CS results.

13. Comparison between the summed ENDF/B-V inelastic CS for all levels from 680 to 1169 keV excitation and the corresponding summed photon CS to the GS rotational band including the β-band 2^+ E0 transition.

14. Inelastic CS from the ^{232}Th β-band 0^+, 2^+ (doublet with 3^-), and 4^+ levels measured by SUNI. The open points with error bars were directly measured, whereas the full points without error bars were deduced from (n;n',γ) results. The authors attribute much of the discrepancy between the two methods to E0 transitions.

15. Inelastic CS to the β-band 2^+ level from ^{238}U (n;n',γ) measurements. The Lowell CS (triangles) is simply the sum of the photon CS for the three E2 decays to the GS band. Although the ORELA summed photon CS (circles without error bars) is similar, the inelastic CS (circles with error bars) is much larger since the measured E0 transition strength of McGowen *et al.* have been incorporated into the decay scheme.

16. Photon spectra from a white-source ^{238}U (n;n',γ) measurement. The hatched area on the lower spectrum is the expect full-energy peak area from prompt fission photons. The hatched peaks coincide with known fission photon isomers and appear at lower neutron energies where they cannot be from inelastic scattering.

17. Inelastic scattering CS to the K=0 octupole band with (data points with error bars) and without (data points without error bars) feeding corrections from higher levels. The calculated curves to 2.0 MeV which reproduce both the shapes and the magnitudes are from Jary *et al.* The curves to 5.0 MeV which overpredict the CS maxima are from the ENDF/B-V evaluation.

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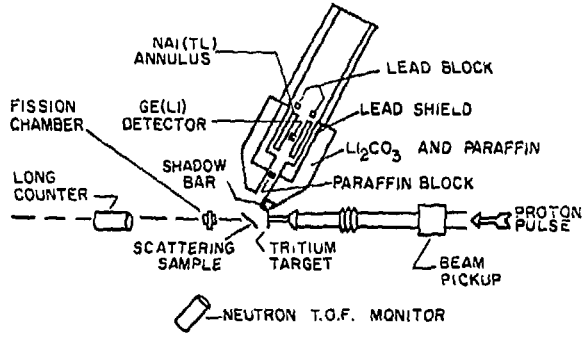

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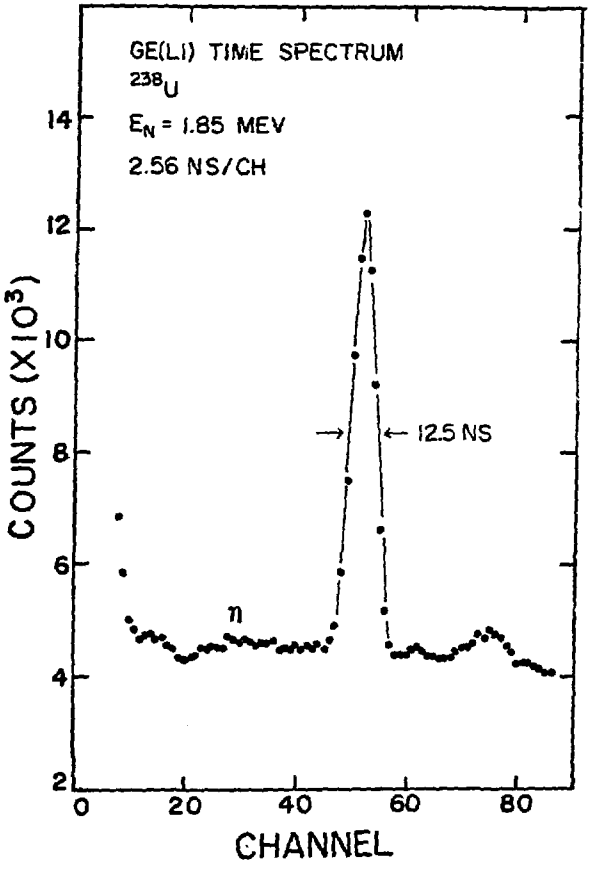

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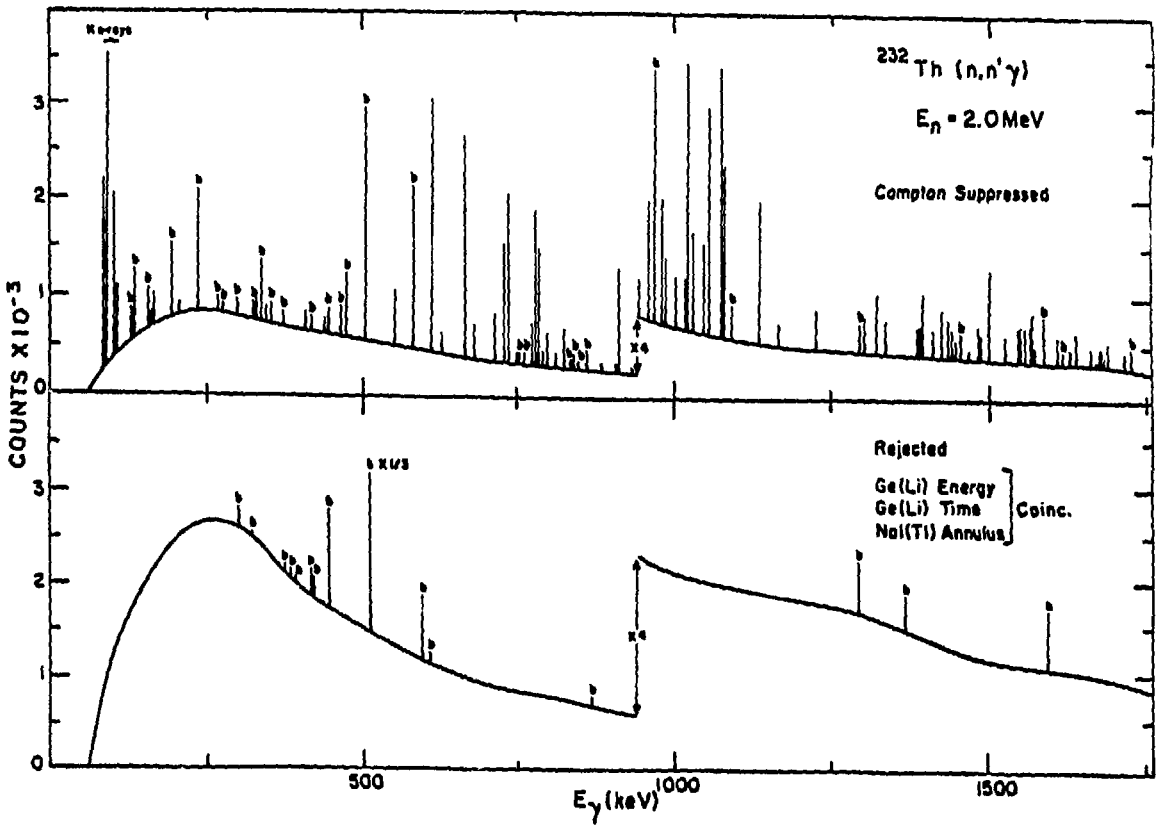

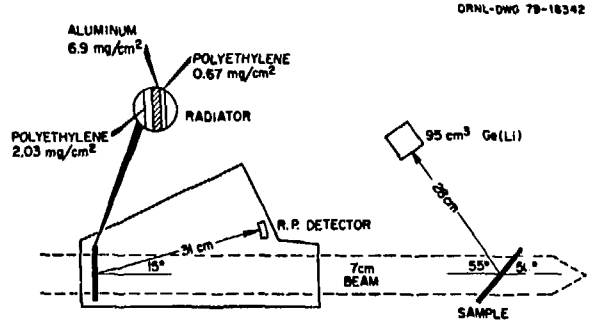
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ALUMINUM
6.9 mg/cm²
POLYETHYLENE
0.67 mg/cm²
RADIATOR
POLYETHYLENE
2.03 mg/cm²
95 cm³ Ge(Li)
28 cm
R.P. DETECTOR
31 cm
15°
7cm
BEAM
55°
SAMPLE

ORNL-DWG. 78-11945

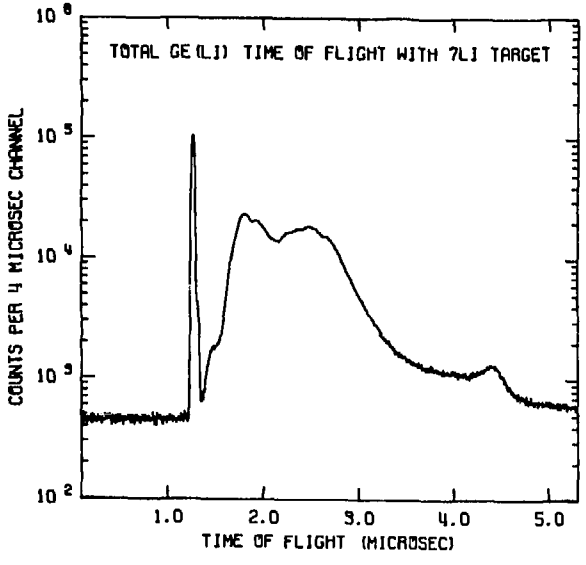
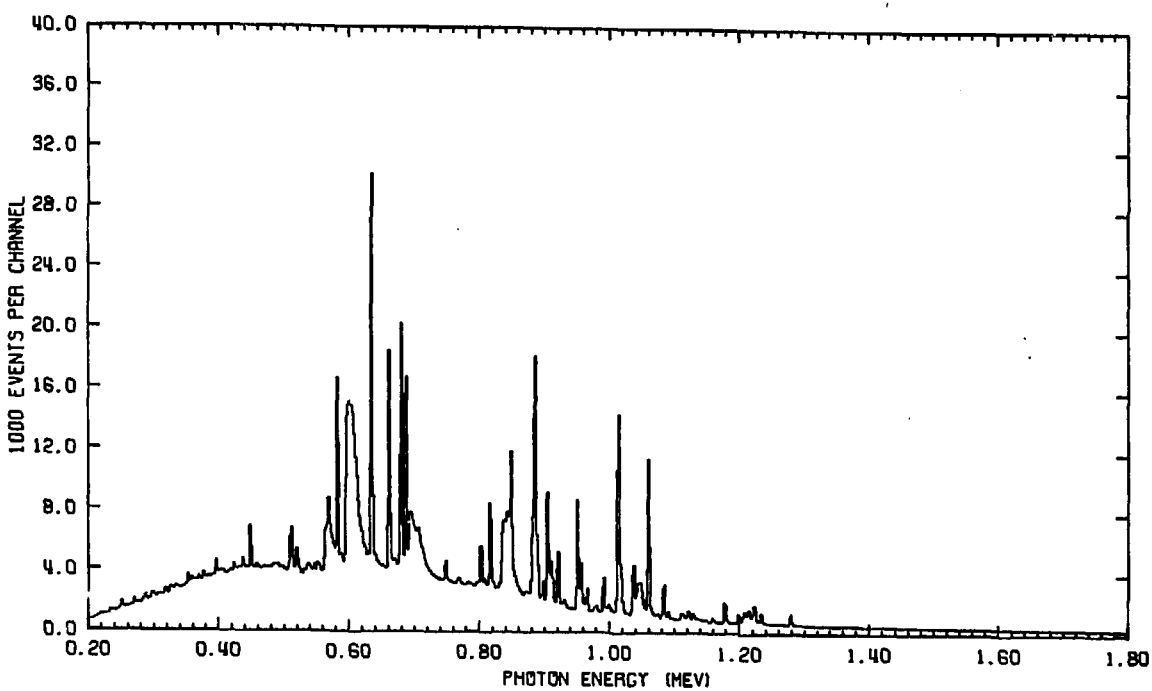


Fig 6

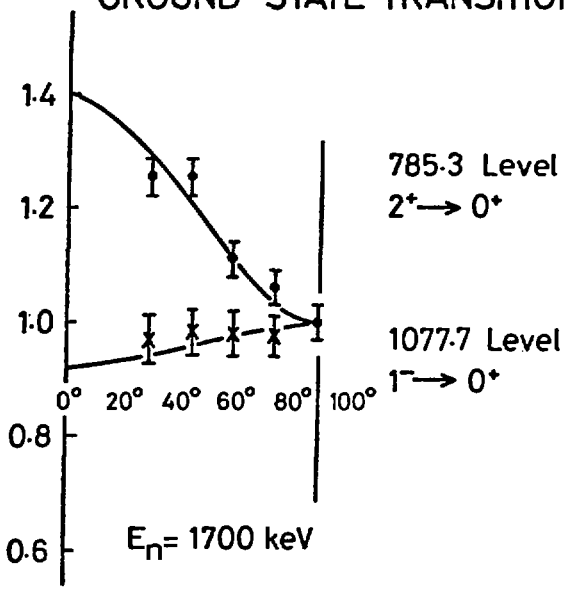
ORNL-DWG 79-9192



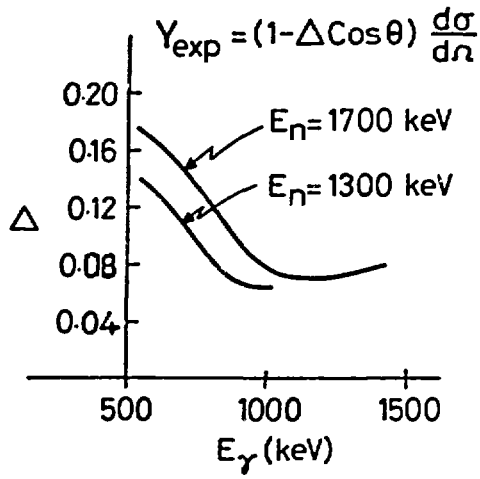
ORNL-DWG 81-21445

GAMMA ANGULAR DISTRIBUTIONS
THORIUM (N,N'γ)

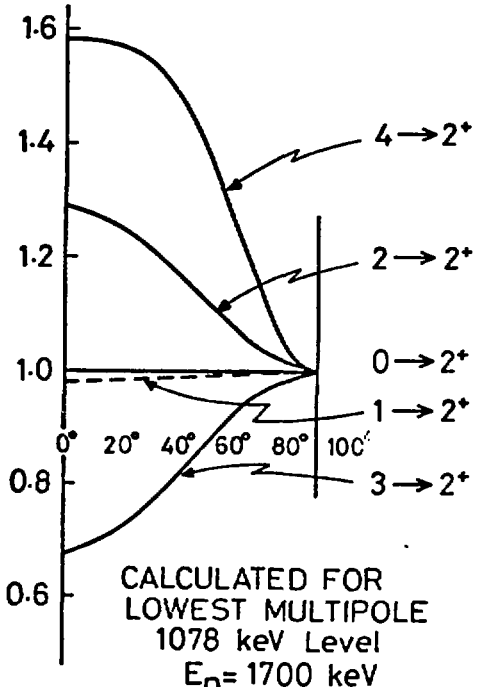
GROUND STATE TRANSITIONS

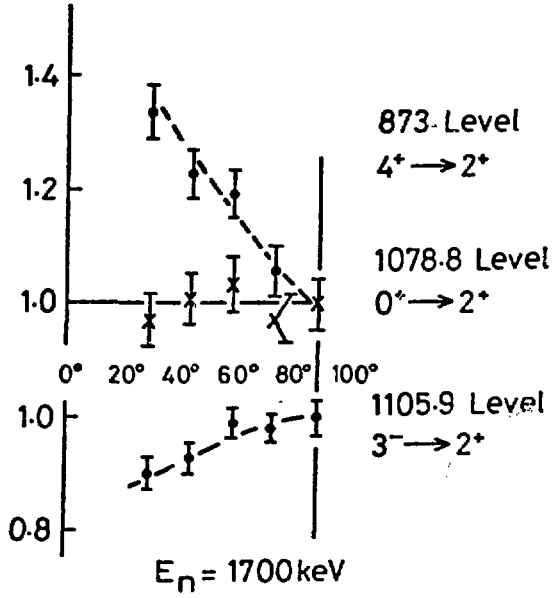


Skewing Correction



TRANSITIONS TO 2⁺ LEVEL AT 49.5 keV





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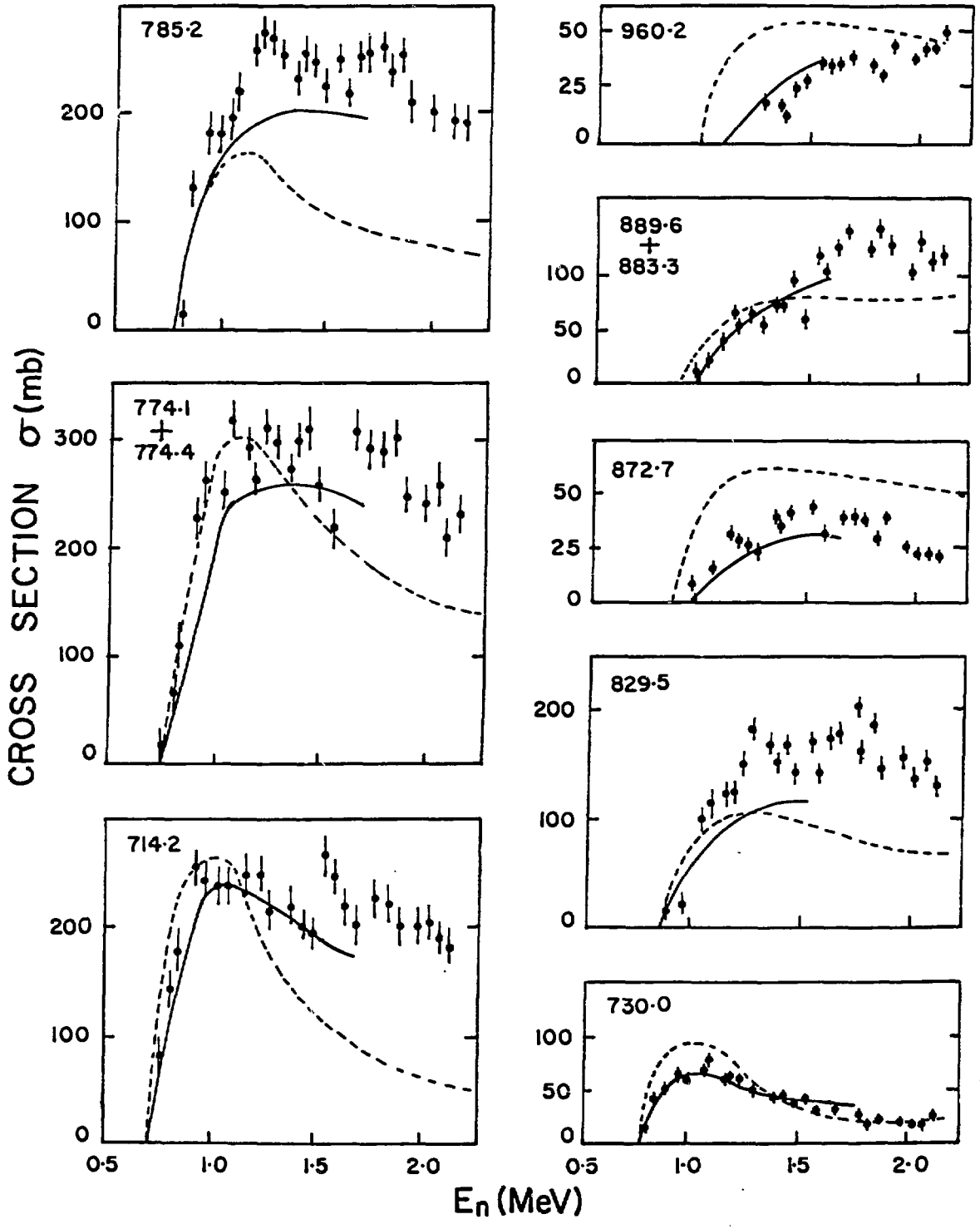

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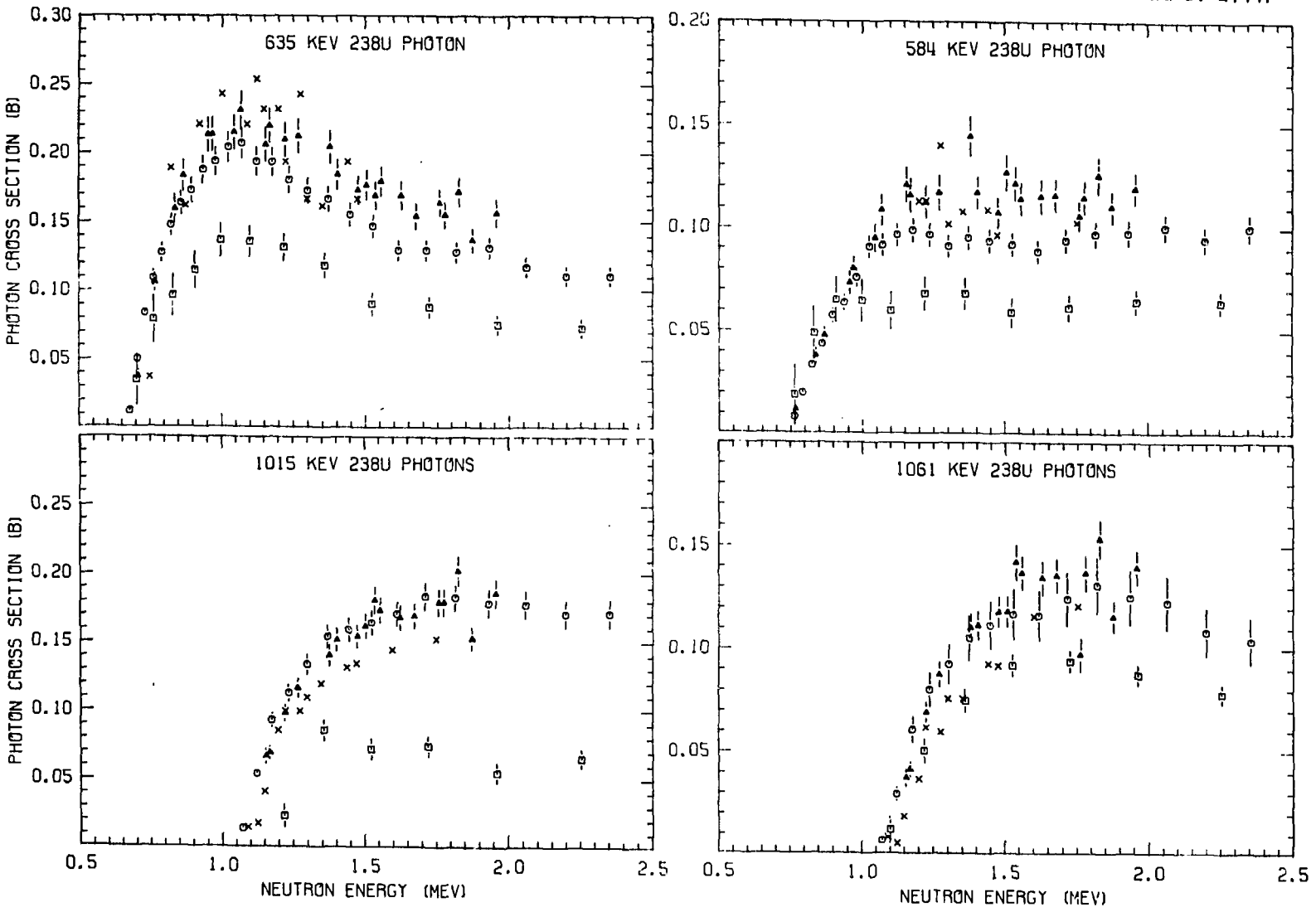

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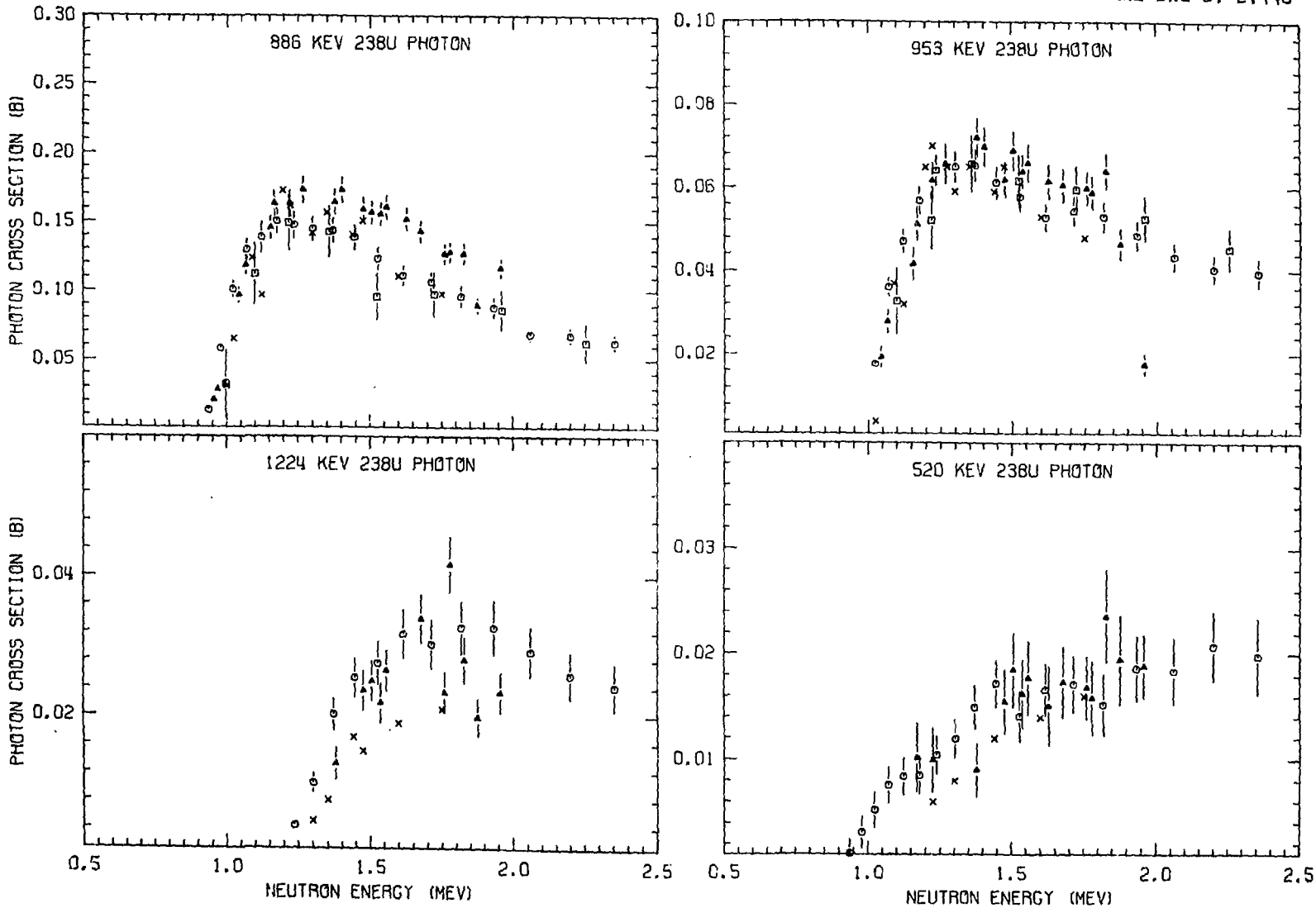

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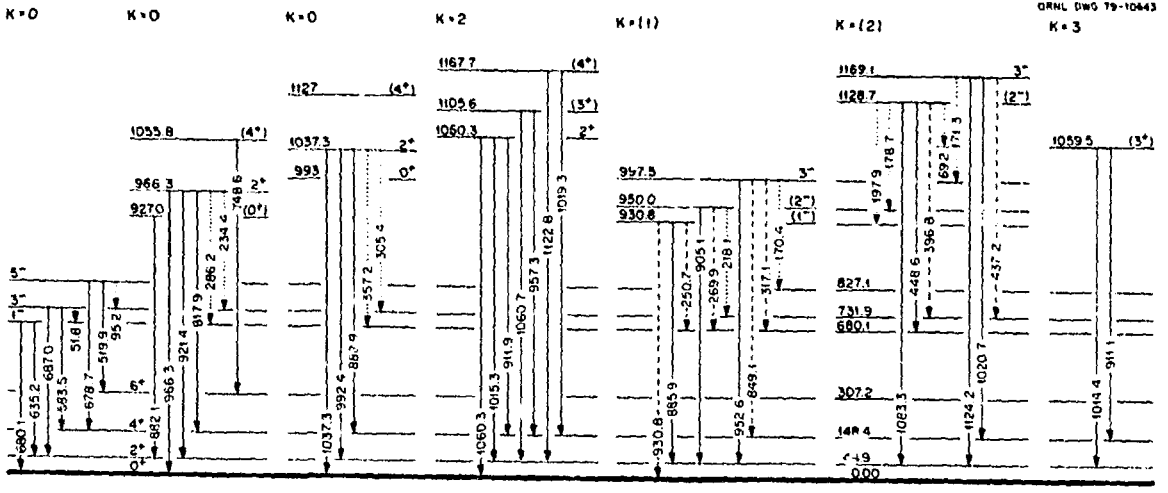

Fig 12

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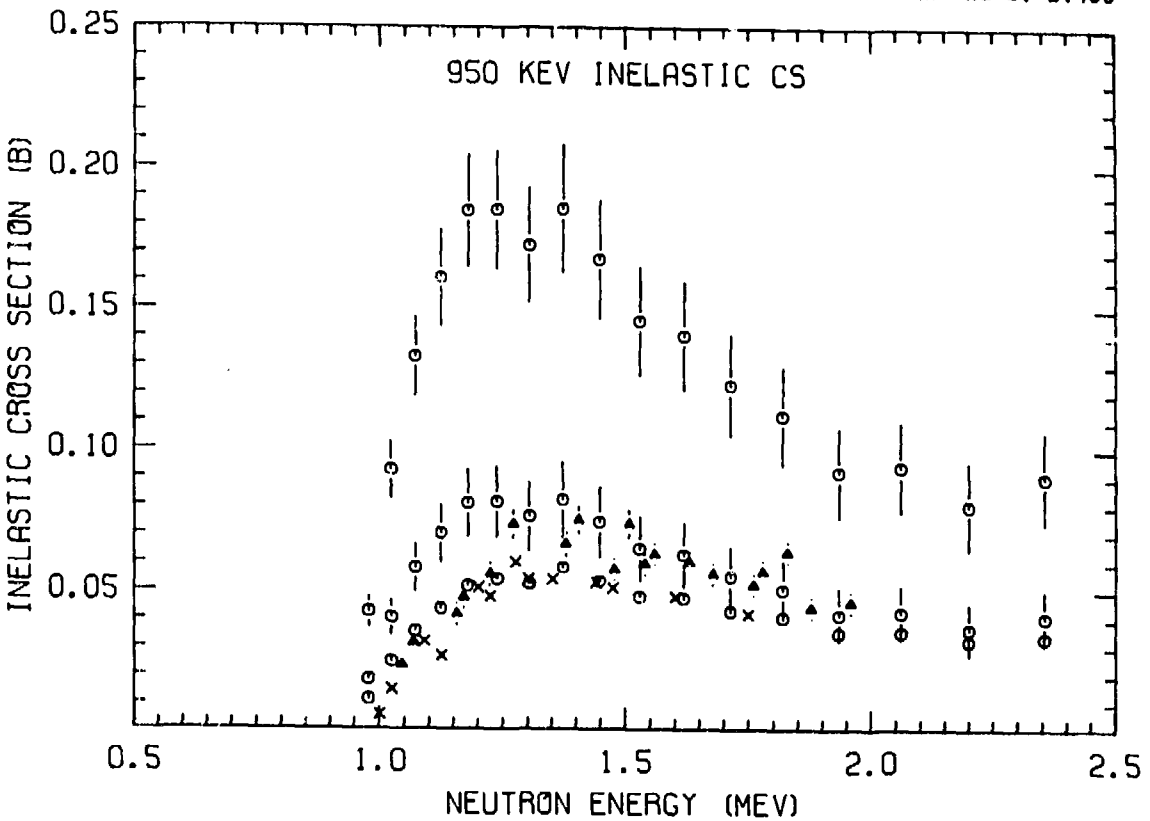
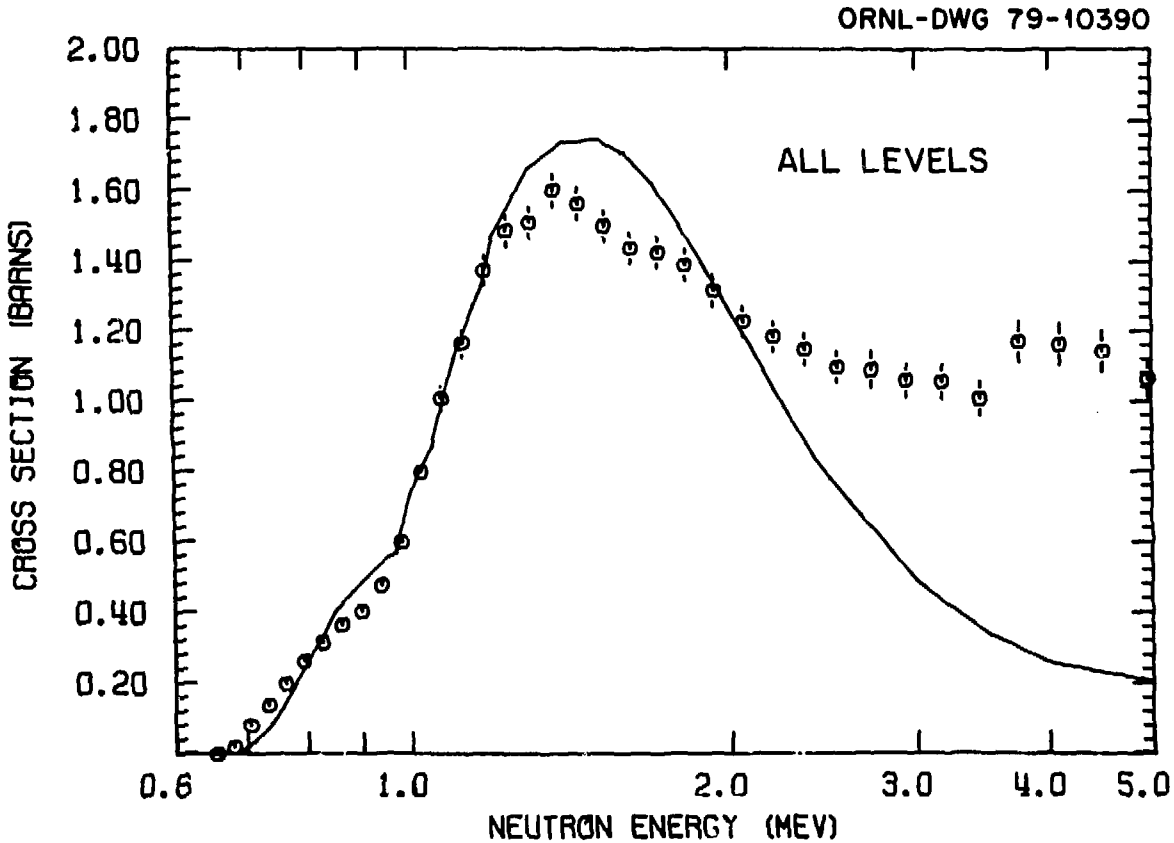



ORNL-DWG 79-10390
ALL LEVELS
CROSS SECTION (BARNS)
2.00
1.80
1.60
1.40
1.20
1.00
0.80
0.60
0.40
0.20
0.6
1.0
2.0
3.0
4.0
5.0
NEUTRON ENERGY (MEV)

ORNL-DWG 81-21451

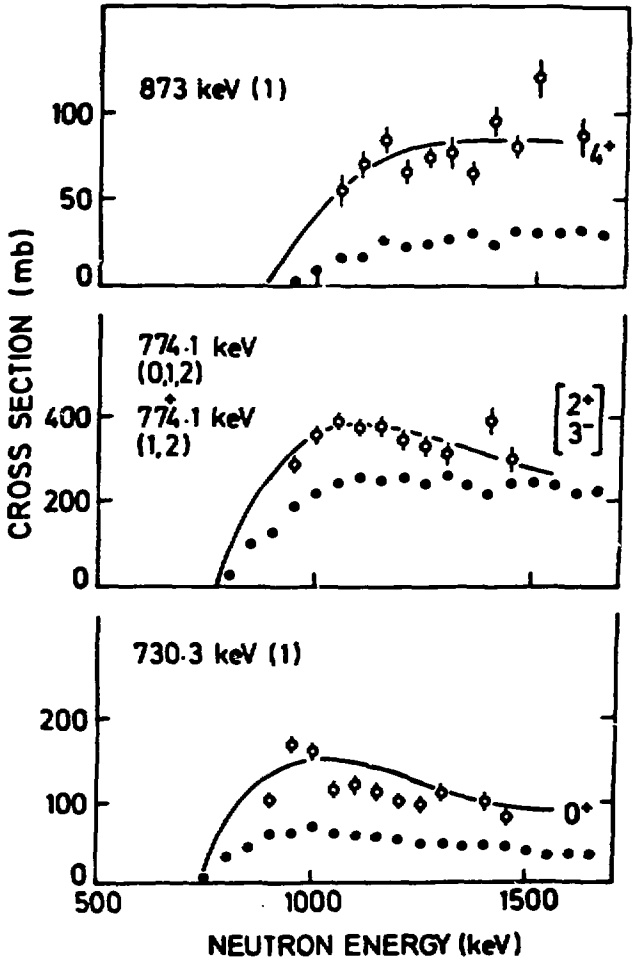
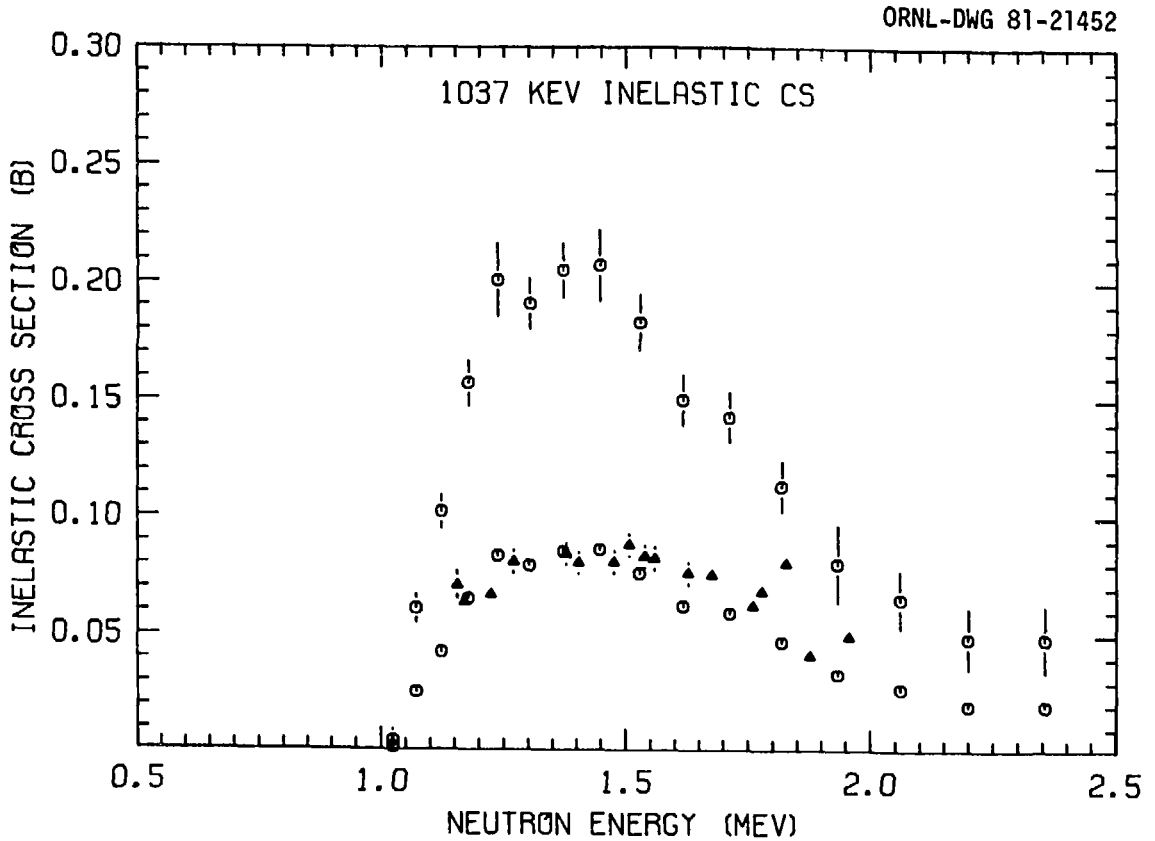
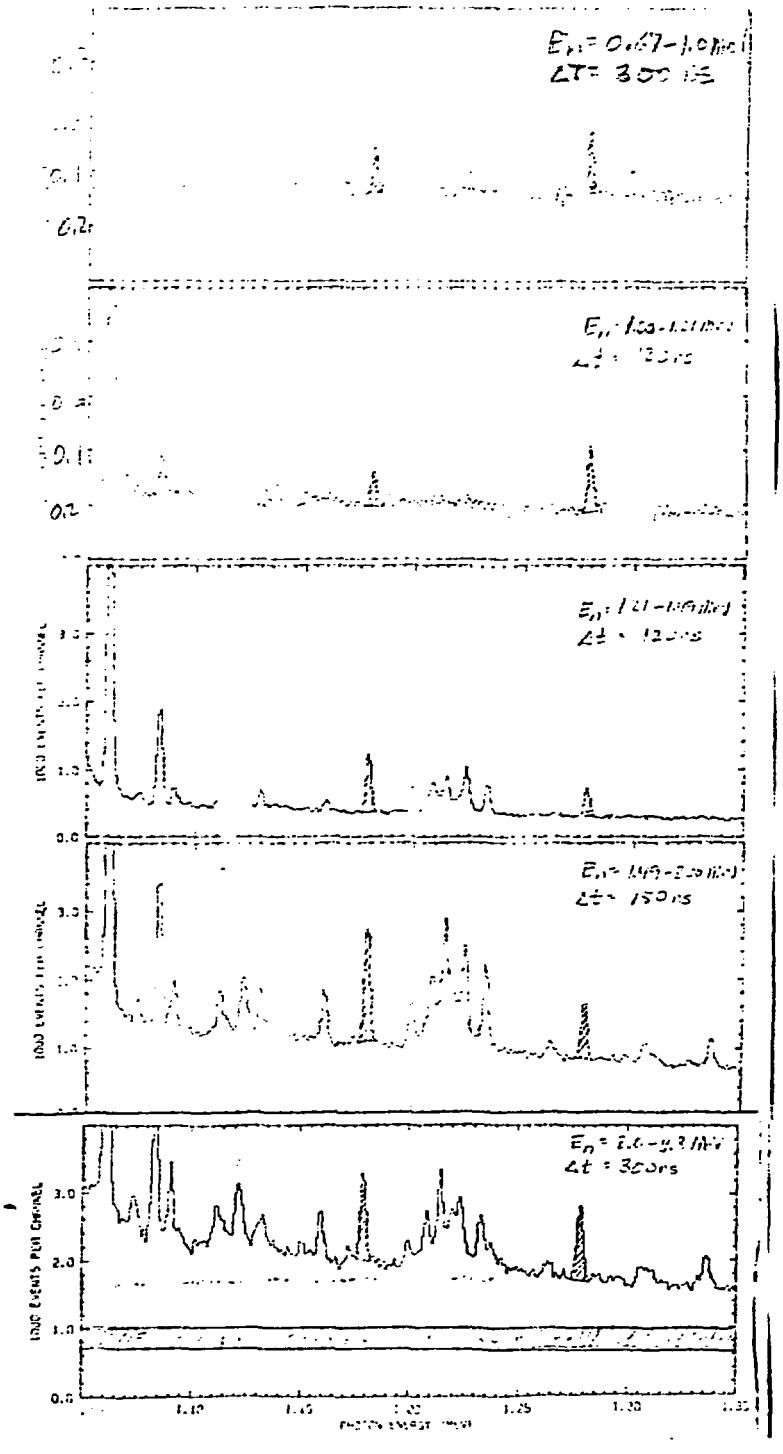


Fig 15


ORNL-DWG 81-21452
1037 KEV INELASTIC CS
INELASTIC CROSS SECTION (B)
0.30
0.25
0.20
0.15
0.10
0.05
0.5
1.0
1.5
2.0
2.5
NEUTRON ENERGY (MEV)

ORNL-DWG-81-21449



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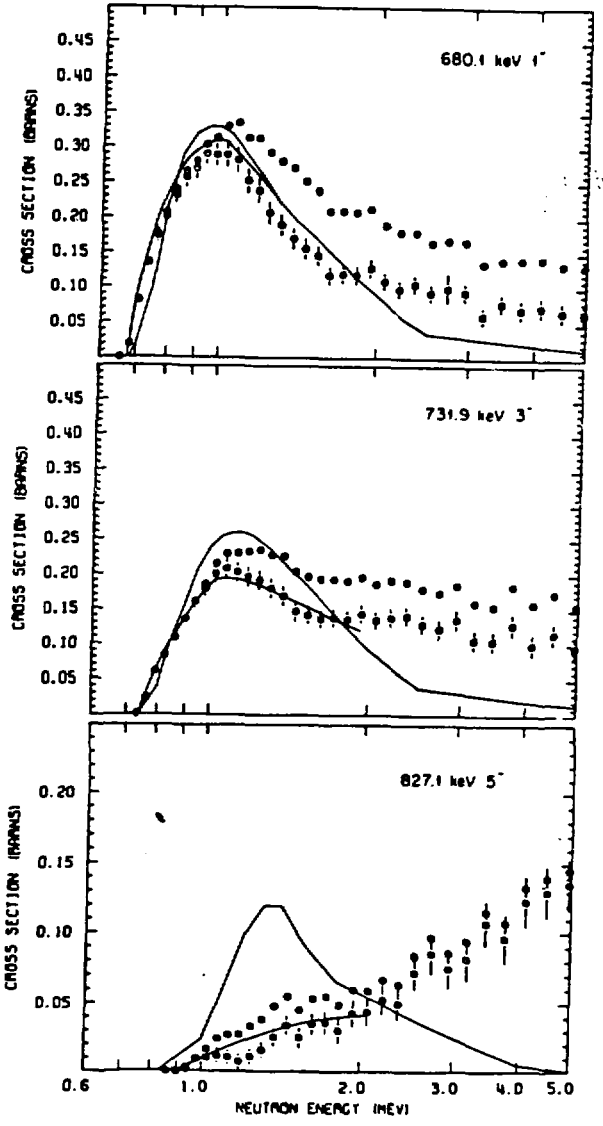
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