NEW EXPERIMENTAL DATA ON ^{238}U NEUTRON INELASTIC SCATTERING

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ABSTRACT

^{238}U neutron inelastic scattering is a source of controversy long since. There are many discrepancies in evaluated data files for partial inelastic cross sections and secondary neutron spectra. The inconsistencies might be attributed, at least partially, to the lack of measurements for incident energies between 2 and 4 MeV.

In this work neutron inelastic scattering cross sections were measured with the time-of-flight technique for incident energies of 2, 2.5, 3 and 3.5 MeV. Differential cross sections were obtained at 35, 55 and 125 degrees for the unresolved levels of the ground-state rotational band and for five groups of levels with excitation energies between 0.63 and 2.68 MeV. Angle-integrated cross sections were obtained for all groups except the ground-state rotational band. The data were measured relative to the elastic scattering cross sections of hydrogen and carbon. The results of the measurements are compared to existing data, existing evaluations, as well as a new model calculation.

A good overall and internally consistent description is obtained for most experimental data with a model that combines the Hauser-Feshbach-Moldauer approach with coupled-channels estimates of elastic scattering and direct reaction contributions. A rigid rotator model describes the ground state band. A soft-deformable rotator was used for the β and γ quadrupole and first octupole bands. The direct contribution to these vibrational bands is essential to describe the experimental data of this work above 3 MeV. The effect on the energy transfer matrix is discussed.

INTRODUCTION

Cross section measurements for neutron inelastic scattering of ^{238}U have been studied often in the last 30 years because these cross sections determine the fast neutron spectrum in nuclear reactors. Nevertheless accuracy requirements (5%) have not been met (Ref. 1). In fact, only in 1989 a NEA working group was started to resolve discrepancies between the major evaluated data files (ENDF/B-VI, JEF-2 and JENDL-3, Ref. 2). Besides the work presented here a considerable number of other measurements were performed recently to aid evaluators in resolving the discrepancies. Simultaneously evaluators have shown that a consistent description may be obtained for most of the available experimental data with an appropriate nuclear reaction model (Ref. 3). The recent experimental data are essential to reduce the ambiguities in the parameters of the model. Below, we demonstrate the ability of this approach to describe our experimental data and the role of our data in fixing model parameters.
Both the total cross section and the transfer matrix (slowing down matrix) of inelastic scattering influence the fast neutron spectrum and, as a consequence k_{eff}, the sodium void coefficient, the antireactivity of the control rod system and the reaction rates. For a PHENIX type fast reactor a sensitivity study gives a 1% change in k_{eff}, a 9% change in the sodium void coefficient, a 5 % change in the Doppler coefficient and a 4% change in the antireactivity as a result of a 20% change in the total inelastic cross section. Given the status of the inelastic cross section this implies that the uncertainties are by no means negligible (Ref. 4). Similarly it was shown that a change in the transfer matrix may result in changes of up to 2% in k_{eff} and, as an example, 20% in the reaction rate ^{238}U-fission/^{239}Pu-fission (Ref. 5). Therefore the neutron emission spectrum is as important as the total inelastic cross section (Ref. 2).
This work contributes to both aspects of inelastic scattering by providing excitation curves for groups of levels between 2 and 3.5 MeV incident energy, where previously few data were available. The present contribution highlights the major aspects of this work and stresses the importance for applications. A full description is in preparation (Ref. 6).

2. EXPERIMENT

Incident neutrons of 2, 2.5, 3 and 3.5 MeV were produced using the T(p,n) reaction. They were emitted from a solid titanium-tritium target of 0.4 mg/cm^2 struck by protons from the pulsed IRMM 7 MV CN Van de Graaff accelerator. A uranium sample was placed on a low-mass Al holder at 12 cm from the neutron-producing target and at zero degrees to the proton beam. The sample was a 3.5 cm diameter by 2 cm high hollow cylinder with an annulus of 1.6 cm diameter. Scattered neutrons were detected by three NE213 detectors at 1.8 m distance from the sample and at nominal scattering angles of 35, 55 and 125 degrees. Each detector was carefully shielded from target neutrons and from neutrons scattered by the other detectors using copper shadow cones and a combination of copper and paraffin collimators. Pulse shape analysis and sample-out measurements were used to eliminate the remaining background components.
The pulsed quasi mono-energetic neutron beam allowed for the measurement of the energy of the emitted neutrons by the time-of-flight technique. The beam pulse width was 1.5 ns with 2.5 MHz repetition rate. The typical energy spread of the incident neutrons was 35 keV.
The measured data were normalized relative to the elastic scattering cross sections of hydrogen and carbon. To this end the scattering yields from a polyethylene and a graphite sample of the

same shape and size as the uranium sample were measured at each incident energy and for each detector. The results of the two normalization methods were averaged and the overall normalization uncertainty is estimated to be 3%.

The detection efficiency of each detector was measured in a separate run. For five incident proton energies the detectors were placed at six angles with respect to the neutron-producing target. Using the known angular distribution of the T(p,n) reaction each proton energy provides an efficiency curve consisting of six points. With sufficient overlap between different proton energies an efficiency curve could be constructed for neutrons of 0.4 to 3.5 MeV with an arbitrary normalization. Pulse height spectra were determined simultaneously. These were fitted with the Monte Carlo code NRESP7 (Ref. 7) to obtain the light-output function characteristic of each detector. Using this light-output function the experimental efficiency curve for each detector is adequately described with the Monte Carlo code NEFF7 and an assumption for the detection threshold. The Monte Carlo code NEFF7 was used to extrapolate the measured efficiency to higher energies and to allow for corrections due to differences in threshold between the scattering measurements and the measurement of the detection efficiency. The estimated contribution of the efficiency to the uncertainty of the differential cross sections is 4-5%.

Throughout the measurements a Pilot-U detector operated in time-of-flight mode served to monitor the time-structure of the proton beam. Two other neutron source monitors served to normalize uranium scattering, sample-out, polyethylene scattering and carbon scattering measurements to unit incident neutron fluence.

Data-analysis consisted of two parts. In the first part background free spectra were obtained using pulse-shape discrimination and the sample-out measurements. These spectra were normalized to a fixed monitor yield. In the second part MCNP (Ref. 8) calculations were performed that included the neutron source characteristics, the samples dimensions and weights, the collimator geometry and the cross sections of the ENDF/B-VI file. The calculated neutron emission spectra take account of the full range of scattering angles, the multiple scattering effects and collimator scattering. They were folded with the time-structure of the proton beam and compared to the experimental data. From a comparison to the hydrogen and carbon scattering measurements the neutron source position and a correction to the detection threshold for each detector were obtained. Following the adjustment of these parameters the normalization factors between calculation (normalized per starting neutron) and experimental data were obtained. The overall agreement between calculated and measured hydrogen (carbon) scattering data was typically 8-14% (5%), depending on detector and incident energy. The normalized uranium neutron emission spectra were subdivided in 6 groups corresponding to the energy of the excited levels (see Table I). For each group the experimental yield was compared to the calculated yield after correction for fission neutrons. The resulting correction factor was applied to the calculated group cross sections as obtained from ENDF/B-VI to obtain the experimental differential scattering cross sections. Overall uncertainties are 10% at 35 degrees and 15% at 55 and 125 degrees. Finally, angle-integrated group cross sections were obtained with an uncertainty of 11%.

Table I. Groups of excitation energies for which differential and angle-integrated data were obtained. The interpretation of the contributing levels in the new model calculation is also indicated.

Group (nr)	Excitation Energy Range (MeV)	Comment
0	0-0.63	Ground state rotational band
1	0.63-0.89	Levels of the first octupole band
2	0.89-1.32	Levels of β, γ and excited octupole bands
3	1.32-1.67	continuum
4	1.67-2.10	continuum
5	2.10-2.68	continuum

3. RESULTS AND DISCUSSION

Figures 1 and 2 show the experimental results for the angle-integrated cross sections of groups 1 and 2 (Table I). The data are compared to measurements by others and to values obtained from the ENDF/B-VI , JENDL-3.2 and CENDL-2.1 data files. Also shown are the results of the new model estimate and their decomposition in compound and direct contributions.

3.1. GROUP 1

For group 1 the new measurements agree well with the trend of the data suggested by the measurements of Aliyar et al., Kornilov et al., Baba et al., Knitter et al. and Shao et al. up to 2.5 MeV (Refs. 9-13). However, at 3.0 MeV the new value is a factor of 2 higher than the value of Aliyar et al. Our cross sections are up to a factor of five lower than those determined by Olsen et al. with the (n,n'γ) technique (Ref. 14). This confirms the results of and Shao et al. and Aliyar et al. It should be noted that for a nucleus like ^{238}U with a high level density at moderate excitation energies, it is very difficult to convert the gamma-ray production cross sections obtained with the (n,n'γ) technique to level or group cross sections. In particular, one needs to know the level and decay schemes accurately and one needs to account carefully for unobserved transitions. Similar discrepancies between results obtained by the (n,n'γ) and (n,n') techniques have been observed for ^{232}Th (Ref. 15).

In Figure 1, two points are observed between 2 and 2.5 MeV that are low compared to other recent measurements. At 2.2 MeV the measurement by Kornilov et al. is low by about 40%. This result was obtained at 120 degree only. Our differential data suggest that an upward correction of about 15 % would result if measurements at other angles were included. The value given by Aliyar et al. at 2.4 MeV is about 50% lower than the angle integrated value that can be obtained from angle differential data also presented in Ref. 9.

Finally, one observes that for group 1 the other authors do not confirm the higher cross section values of early measurements by Barnard et al. and Batchelor et al. in the energy range of 0.8 to 2.0 MeV (Refs. 16 and 17).

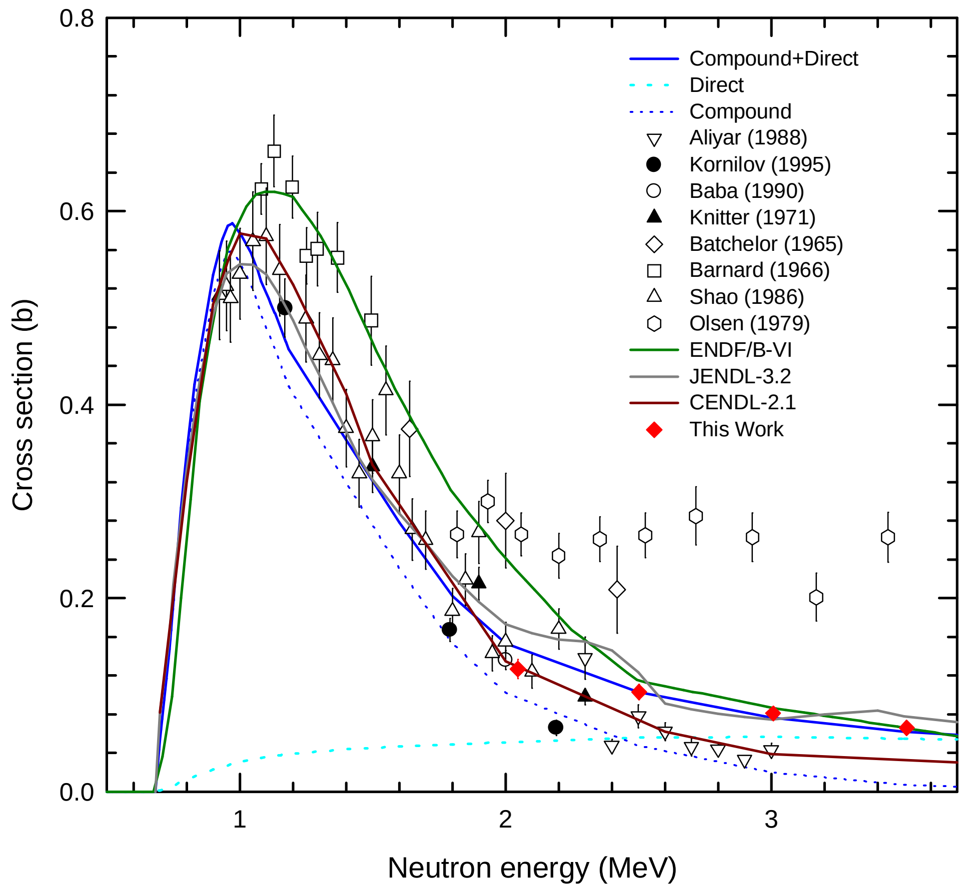

Figure 1. Experimental data for group1 compared to other data and evaluations

The ENDF/B-VI evaluation is seen to agree well with the new data at 2.5, 3.0 and 3.5 MeV. Nevertheless it overpredicts the new value at 2.0 MeV by almost a factor of 2. An overestimate of 20-30% of the measurements by ENDF/B-VI is systematic in the energy range of 0.8 to 2.0 MeV as it fits the early results by Batchelor et al. and Barnard et al.

The JENDL-3.2 evaluation describes the measurements at 2.5, 3.0 and 3.5 MeV equally well and overpredicts the 2.0 MeV data point by 30%. It provides a good description of the data between 0.8 and 2.0 MeV. The peculiar wavy nature of the excitation curve above 2.0 MeV is entirely due to the continuum contribution of the data file.

CENDL-2.1 is an ongoing evaluation that at present fits all the recent data and in particular those of Aliyar et al. It therefore underestimates our new data at 2.5, 3.0 and 3.5 MeV while describing the 2.0 MeV data point well.

The new model description labeled “Compound+Direct” is seen to describe the new data well. Above 2.5 MeV the calculation is dominated by the direct contribution to the excitation of the levels of the first octupole band. Between 0.8 and 2.0 MeV the new calculation slightly underestimates the other experimental data. This is currently under investigation.

3.2 GROUP 2

For group 2 a limited number of other measurements are available (see Figure 2). Only at 2.5 MeV can a direct comparison be made. The point of Baryba et al. (Ref. 18) is about 23% lower than ours. Nevertheless the data of Shao et al., Kornilov et al., Baryba et al. and the new data suggest a clear trend for the excitation curve of this group. Earlier measurements by Batchelor et al., Barnard et al., Cranberg et al. (Ref. 19) and Knitter et al. appear systematically too high below 2.0 MeV incident energy.

ENDF/B-VI overestimates the new data considerably (up to a factor of 2) and seems to ignore the trend offered by the experimental data.
JENDL-3.2 offers a reasonable description of the present data at 3.0 and 3.5 MeV, whereas it generally overpredicts all recent data between 1.2 and 2.5 MeV. In particular, it seems to aim at describing the earlier measurements.

The model calculation "Compound+Direct" describes the trend of the recent data from 1.2 to 3.5 MeV rather well, although there is a slight overprediction. In this case the direct contribution to the excitation of the β and γ quadrupole bands dominates the description of our data at 3.0 and 3.5 MeV.

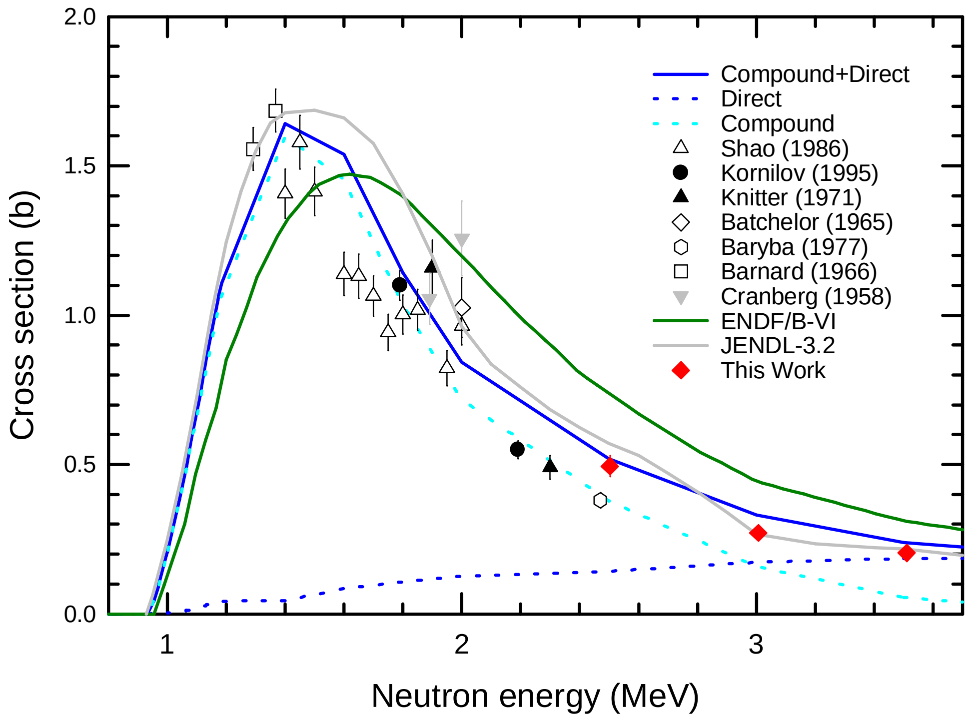

Figure 2. Experimental data for group2 compared to other data and evaluations

3.3 THE MODEL DESCRIPTION

The present description is an improvement over those presented in Ref. 3 and the thesis of C. Goddio (Ref. 6). The coupled channels estimate of the direct contribution was tailored to describe the new experimental data. This is feasible, since the deformation parameters of the soft rotator model were fitted both to scattering data and excited level scheme. It should be stressed that the model estimates also use the proper description of the experimental total cross sections and the angular distributions of the ground state rotational band and the excited levels as a constraint. In addition, a fit is made to the fission cross sections by appropriate modeling of the level densities of $^{238,239}U$ and the use of a double-humped fission barrier. All these aspects are included in a consistent model approach avoiding arbitrary renormalization of the calculated data to obtain a better fit.

The total inelastic scattering cross sections obtained from the model calculations closely follow those of JENDL-3.2 leading to values about 5% larger than those of ENDF/B-VI from 1 to 3 MeV incident energy. Above 3 MeV the situation is reversed and at 5 MeV the difference amounts to 9%.

As indicated earlier further improvements should be sought and are feasible to obtain a good agreement with all recent experimental data.

4. CONCLUSION

New experimental data have been obtained for inelastic neutron scattering by ^{238}U for incident energies of 2.0, 2.5, 3.0 and 3.5 MeV. Differential cross sections were obtained at 35, 55 and 125 degrees for five groups of levels with excitation energies between 0.63 and 2.68 MeV and for the unresolved levels of the ground state rotational bands. Angle integrated cross sections have been obtained for the five groups of excited levels with an uncertainty of 11%. The angle integrated results for groups 1 and 2 have been presented here and compared to existing data and the major evaluations. It was shown that a complete excitation curve is offered by recent measurements with few remaining inconsistencies. The new measurements offer only the second set of (n,n') data in the high energy tail of these curves. ENDF/B-VI does not offer a good description of these excitation curves, as it was not adjusted to any of the more recent data. JENDL-3.2 is significantly better for group 1 but should be improved for group 2 and for the emission of neutrons by continuum levels.

It was shown that a consistent model that offers a fair description of all neutron induced cross sections, also describes our data well by fine-tuning the deformation parameters governing the excitation of the first octupole and the β and γ quadrupole vibrational bands.

A full publication of all our results is in preparation. Nevertheless it is of interest to mention that a softer spectrum would evidently result if ENDF/B-VI were adjusted to provide a better description of the data. Also this is the case for JENDL-3.2 in the case of groups 3-5. Finally, it may be noted that a full revision of the description of ^{238}U neutron scattering data to obtain the best description possible within the model's constraints is subject of an ISTC proposal.

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