# NIST Measurements and Standards Related Work at Other Facilities

### Allan D. Carlson Neutron Physics Group National Institute of Standards & Technology

Presented at The CSEWG Meeting BNL November 8, 2012

#### THE NEUTRON CROSS SECTION STANDARDS

Reaction	Energy Range
H(n,n)	1 keV to 20 MeV
<sup>3</sup> He(n,p)	thermal to 50 keV
<sup>6</sup> Li(n,t)	thermal to 1 MeV
<sup>10</sup> B(n,α)	thermal to 1 MeV
$^{10}\mathrm{B}(\mathrm{n},\alpha_{1}\gamma)$	thermal to 1 MeV
C(n,n)	thermal to 1.8 MeV
$^{197}$ Au(n, $\gamma$ )	thermal, 0.2 to 2.5 MeV
<sup>235</sup> U(n,f)	thermal, 0.15 to 200 MeV
<sup>238</sup> U(n,f)	2 to 200 MeV

NIST National Institute of Standards and Technology • Technology Administration • U.S. Department of Commerce

### H(n,n)H Angular Distribution Measurements

•There is a problem with the quality of data at small CMS angles for hydrogen scattering. In order to improve the database of measurements at smaller scattering angles an experiment has been designed where the primary objective is detection of the scattered neutron instead of the scattered proton.

•The work is being done at the Ohio University accelerator facility. Preliminary measurements have been made at laboratory neutron scattering angles from 20 degrees to 65 degrees in 5 degree steps for 14.9 MeV incident neutrons. The plan is to increase the accuracy of the measurements and extend the angular range so that data are obtained from 15 to 70 degrees.

•To obtain the accuracy needed for this work, the neutron detector efficiency must be determined accurately. At neutron energies below about 9 MeV <sup>252</sup>Cf spectra are being used.

### H(n,n)H Angular Distribution Measurements (cont.)

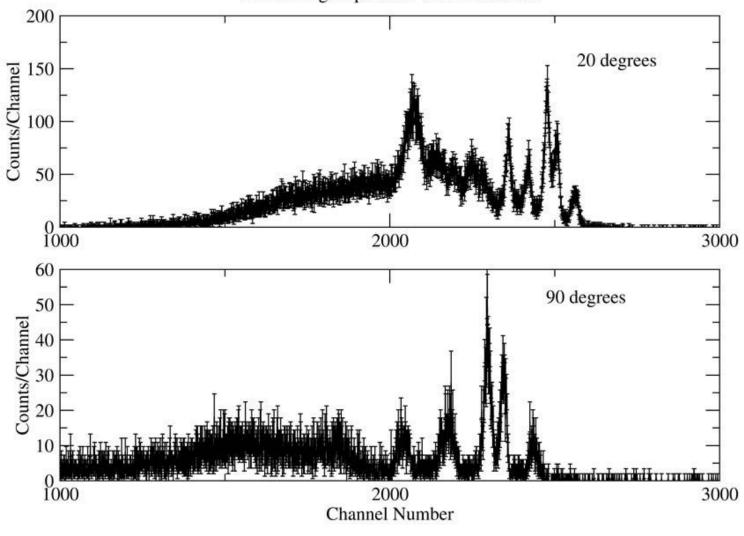
•For neutron energies above 9 MeV, a technique using reactions where the projectile and target are identical is being used. Because they are identical, the angular distribution **must** be symmetrical in the CMS. So the neutron yield at an angle  $\Theta$  must be the same as that at 180-  $\Theta$  in the CMS. But the energies of the neutrons are different in the LAB system. Thus in the LAB system, for a bombarding energy such that the backward portion of the angular distribution falls in the energy range below 9 MeV where the efficiencies are well known, we can deduce the efficiency for the higher energy group in the forward hemisphere. Analysis of the measured counts at the appropriate angles can then give us the efficiency for the 14 MeV neutron energy range.

Our study indicated that the  ${}^{6}Li({}^{6}Li,n){}^{11}C$  reaction would be the best for our use, however for the only suitable targets we successfully made,  ${}^{6}LiF$  and  ${}^{6}LiCl$ , the backgrounds were very large. Measurements with C(C,n) were successful. Additional work is planned with the D(d,n) and  ${}^{13}C({}^{13}C,n)$  reactions. This is an ongoing project.

(collaboration of Ohio University, NIST, LANL and the University of Guelma)



#### First Measurements of neutrons from the C(C,n) Reaction



Time of flight spectrum with neutron cut

NIST National Institute of Standards and Technology • Technology Administration • U.S. Department of Commerce

### H(n,n)H Angular Distribution Measurements (cont.)

•Additional work on the angular distribution is planned at PTB by Nolte and Schmidt. With the neutron detector array they have, they will make measurements at 5 angles simultaneously avoiding normalization of the detectors. 10 MeV and 14 MeV measurements are under consideration. Measurements could be made at the smaller CM angles: 25°- 80° for 10 MeV and to about 65° for 14 Mev. The limiting factor is they will not be using an active sample (detector) so scattering from carbon in the sample is a problem. They will normalize the data with a proton recoil telescope at 0° in the lab. They are concerned about getting accurate determination of the hydrogen content in the acetanilide sample being used.

### H(n,n)H Angular Distribution Measurements (cont.)

•Plans are being made to make hydrogen angular distribution measurements using a Time Projection Chamber which will provide higher counting rates than are possible with the other methods.

(collaboration of Ohio University and LANL)

## **Hydrogen Angular Distribution at High Neutron Energies**

•The most recent measurements of the hydrogen angular distribution in the 100 MeV energy region are not consistent at back angles. Larger cross sections were measured at Uppsala (96 and 162 MeV) and PSI (many energies from about 280 MeV to 580 MeV), both using pseudo-monoenergetic sources. The work at Indiana University at 194 MeV, using neutrons tagged by detection of the associated protons from the D(p,n)2p reaction, indicate lower cross sections and they agree with PWA calculations.

•The Uppsala group suggest that the Indiana experiment may be preferred due to the smaller total uncertainties.

•Though there is an indication that the discrepancy may be resolved at about 160 MeV - 200 MeV, the PSI data which cover a very large energy range (200-580 MeV) still stand as measured. Further work should be done to understand this problem.

•Also more work should be done on angular distribution measurements in the intermediate energy region from about 30 MeV to 150 MeV. Little data are now available and the angular interval measured is very limited.

## <sup>3</sup>He(n,p) Measurements

•Progress continues on an experiment to measure the n-<sup>3</sup>H coherent scattering length. This measurement is complementary to the n-<sup>3</sup>He work. This measurement would constrain the fundamental nucleon-nucleon interaction models that underlie all of our cross section work. This work could help with Hale's R-matrix evaluation of the <sup>3</sup>He(n,p) cross section.

(collaboration of NIST with Indiana University and the University of North Carolina)

## <sup>6</sup>Li(n,t) Measurements

• A measurement has been completed of the  ${}^{6}\text{Li}(n,t)$  cross section standard at ~ 4 meV neutron energy. This is the first direct and absolute measurement of this cross sections in this neutron energy range using monoenergetic neutrons. A primary effort was very accurate measurements of the fluence. The fluence (efficiency) has now been determined with an uncertainty of 0.05%.

•The limitation on the accuracy of the <sup>6</sup>Li(n,t) cross section measurement is the mass uncertainty of the <sup>6</sup>Li target. The present mass uncertainty is about 0.25%. The deposits were made at IRMM. Studies have been made to compare the mass with the value obtained when it was characterized a number of years ago. Comparisons have also been made with a number of other deposits made at the same time at IRMM. It is expected that an ultimate total uncertainty less than 0.3% for the cross section can be obtained from this experiment.

(collaboration of NIST, LANL, the University of Tennessee and Tulane University)

### <sup>6</sup>Li(n,t) Measurements (cont.)

• Work is underway at IRMM on <sup>6</sup>Li(n,t) measurements.

•At the GELINA linac, Hambsch plans angular distribution and cross section measurements for the <sup>6</sup>Li(n,t) reaction. The cross section data will be relative to the <sup>235</sup>U(n,f) standard. This work will extend from a few keV to about 3 MeV so the resonances at 0.25 and the weak one at about 2 MeV will be covered. The <sup>6</sup>Li samples were supposed to be made by the end of October. They are using a digital data acquisition system for these experiments. Measurements should start this year.

•At the IRMM Van De Graaff facility Giorginis and Bencardino are begun making <sup>6</sup>Li(n,t) measurements at the IRMM Van De Graaff facility. They are using a Time Projection Chamber that was designed and fabricated at IRMM. They plan to obtain high cross section accuracy by determining all important parameters with the best possible precision: number of reaction events, number of monitor events, number of <sup>6</sup>Li atoms in the <sup>6</sup>LiF samples, and the number of <sup>238</sup>U atoms in the monitor. They are characterizing the <sup>6</sup>LiF samples with Thermal Neutron Depth Profiling. They will be obtaining <sup>6</sup>Li(n,t) cross section data in the 2 MeV energy region. These data should overlap the GELINA data from 1 to 3 MeV. The <sup>6</sup>Li deposits for both the GELINA and Van De Graaff measurements were made at IRMM.

### <sup>10</sup>B(n, a) Measurements

•Hambsch continues to accumulate data on the branching ratio, the angular distribution and the <sup>10</sup>B(n, $\alpha$ ) and <sup>10</sup>B(n, $\alpha_1\gamma$ ) cross sections relative to the <sup>235</sup>U(n,f) standard up to about 3 MeV. This work is being done at the 60m station of GELINA at IRMM. He found problems with the branching ratio data from 1 – 2 MeV that he published in Nucl. Sci. Eng. 156 (2007) 111. He has taken improved data with very good statistics that cover that energy region. The new branching ratio measurements in the 1-2 MeV energy region are now in better agreement with the ENDF/B-VI and ENDF/B-VII evaluations. That are presently analyzing the newly measured data.

### <sup>10</sup>B(n,a) Measurements (cont.)

• The Zhang et al. measurements will have application for the standards when the energy range is increased above 1 MeV. Their Frisch gridded ionization chamber work on the <sup>10</sup>B(n, $\alpha$ ) angular distribution relative to the <sup>238</sup>U(n,f) standard at 4 and 5 MeV is not affected by "particle leaking". These data are in very good agreement with the 2006 data of Giorginis and Khryachkov. However they communicate that there is a "problem" with the data at 4.17, 5.02, 5.74, and 6.52 MeV, apart from "particle leaking." This is being investigated.

• They also have found that there appears to be a loss of <sup>10</sup>B from their samples as a function of time. They found that the sample suffers almost a 40% loss in thicknesses in about 2 years' duration. They do not know if the loss is related to neutron beam bombardment. As a result of these problems, they are doing a systematic study of their work. His latest communication indicated they are still working on this problem.

•NIST has also noted a problem with loss of <sup>10</sup>B from evaporated deposits. NIST plans for making <sup>10</sup>B(n, $\alpha$ ) measurements at low neutron energies are being delayed while work is being done to investigate these losses.

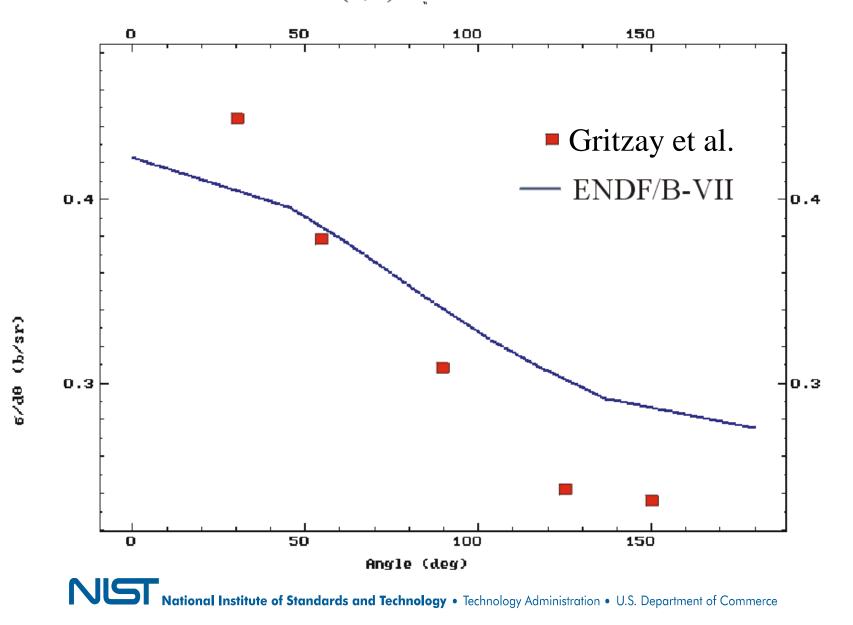
## C(n,n) data

• Gritzay et al. data were shown at the last CSEWG Meeting. they have angular distribution data for 2, 59 and 133 keV. The data were taken at the Kyiv reactor using filered beams. The measurements were made at 30, 55, 90, 125 and 150°. They were measured relative to lead scattering but the shape should still be relatively good anyway.

In her last correspondence she indicated she was planning on continuing this work. But her immediate effort is developing a new method for measuring the carbon total cross section in the 90 - 160 keV energy region that encompasses the 152.9 keV carbon resonance. Her previous work indicated that the resonance has a much larger neutron width than previously thought. The method she is using is called modified filtered beams. It involves making measurements with many different types of filters and analyzing the results. A paper on this work was given at the NPAE-2012 conference in Kiev.

The measurements at 133 keV are shown in the next figure. The results differ from the carbon standard.

 $C(n,n) \ge 133 \text{ keV}$ 



### Au $(n,\gamma)$ at low energies

• To support the needs of certain applications, such as astrophysics, the energy range below about 100 keV for gold capture haas been added to the standards activities as a "reference" cross section.

• Due to the evaluation process used for the standards evaluation, data for the Au $(n,\gamma)$  cross section were obtained for energies below 200 keV.

• These results are consistently higher than the Ratynski evaluation (by about 5-7% from 15 to 25 keV) which is used in astrophysics applications.

• The Ratynski evaluation relies on Macklin capture data and Ratynski-Käppeler Karlsruhe pseudo-Maxwellian capture data.

• The standards evaluation uses a large database of various types of data.

### Au $(n,\gamma)$ at low energies (cont.)

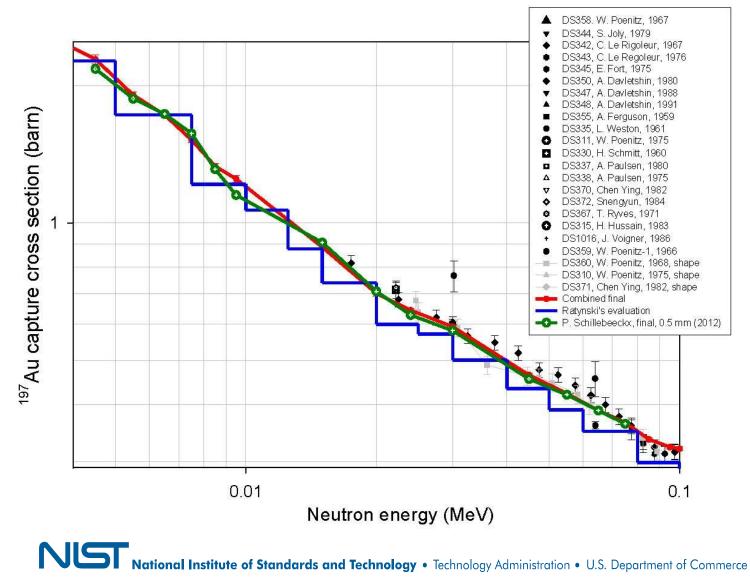
• New experiments on the  ${}^{197}Au(n,\gamma)$  cross section

•Wallner et al. using AMS with samples irradiated in a simulated Maxwellian neutron source spectrum of 25 keV mean energy obtained a ratio to the standards evaluation for gold capture of  $1.05 \pm 0.06$  (obtained using his  $^{238}U(n,\gamma)/Au(n,\gamma)$  cross section ratio and the  $^{238}U(n,\gamma)$  cross section from the standards evaluation)

•Lederer et al. reanalyzed n\_TOF gold capture data of Massimi et al. and folded a simulated Maxwellian neutron source spectrum of 25 keV mean energy into that data. It agrees with the standards evaluation. The result with an uncertainty of 3.6% is smaller than the standards evaluation by 1%. It is 4.7% higher than the Ratynski evaluation.

•GELINA Au $(n,\gamma)$  cross section measurements by Borella et al., Lampoudis et al. and Schillebeeckx et al. all support the standards evaluation.

#### Low energy Au $(n,\gamma)$ cross section measurements and evaluations



### Au $(n,\gamma)$ at low energies (cont.)

• New experiments on the  ${}^{197}Au(n,\gamma)$  cross section (cont.)

•Recent measurements have been made by Feinberg et al. of the 197Au( $n,\gamma$ ) cross section using <sup>7</sup>Li(p,n)<sup>7</sup>Be neutrons. They made the measurements with a proton energy of 1912 keV at IRMM. Data were obtained using proton beam energy spreads of 1.5 keV and about 20 keV. Both measurements were about 5% higher than the Ratynski evaluation and the Macklin measurements. However the difference is only about  $2\sigma$ . The measured values were compared with detailed calculations obtained with simulated spectra folded into the ENDF/B-VII.0 cross sections. The agreement was within 2%.

### Au $(n,\gamma)$ and <sup>238</sup>U $(n,\gamma)$ Measurements

• In addition to the measurements at 25 keV by Wallner (U. of Vienna) of the  ${}^{238}U(n,\gamma)/{}^{197}Au(n,\gamma)$  cross section ratio, neutron irradiation data were obtained for this ratio at 426 keV. Accelerator mass spectrometry was used to measure the  ${}^{239}$ Pu resulting from the  ${}^{239}$ U. Activation was used for the gold measurements. The measurement has a large (150 keV FWHM) energy spread. That ratio,  $0.99 \pm 0.04$ , compared with the standards evaluation is in excellent agreement.

## <sup>238</sup>U(n, γ) Measurements

• Ullmann et al. made measurements of the  ${}^{238}$ U(n, $\gamma$ ) cross sections using the DANCE (160 BaF<sub>2</sub> crystals) detector at LANSCE. The neutron beam was monitored with a  ${}^{235}$ U fission chamber, a BF<sub>3</sub> counter, a  ${}^{6}$ Li F detector and a  ${}^{3}$ He detector. Small  ${}^{238}$ U samples could be used due to the high neutron intensity at DANCE. This reduces the uncertainty due to multiple scattering. Though the data could be made absolute, they normalized to capture in the 80 and 145 eV resonances. They associate a 2 percent uncertainty to this normalization. They state there is generally good agreement with the ENDF/B-VII evaluation. He is presently working on the normalization of that data and expects results by the end of the year.

## <sup>238</sup>U(n,γ) Measurements (cont.)

• Measurements have been made recently at GELINA and n\_TOF using the same sample.

•At GELINA, Lampoudis et al. made measurements using a  $C_6D_6$  detector. The data are now being analyzed.

•At n\_TOF, measurements were made with a  $C_6D_6$  detector by Mingrone et al. and with a BaF<sub>2</sub> detector by Wright. They are analyzing the data. They anticipate that average values will be available from 20 keV to 1MeV. Below 20 keV resonance data will be available.

•It is anticipated that results of all these measurements will be given at the ND2013 conference. Abstracts for the presentations have been accepted.

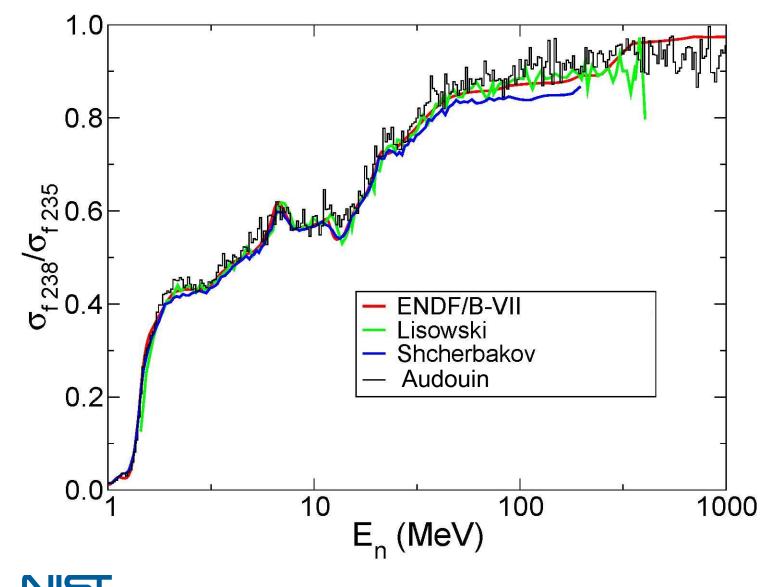
### <sup>235, 238</sup>U(n,f) and <sup>239</sup>Pu(n,f) Measurements

•There has been no new measurement activity since the last CSEWG meeting for these fission cross sections. The data sets from the two independent measurements of the  ${}^{238}\text{U}(n,f)/{}^{235}\text{U}(n,f)$  cross section ratio by Calviani et al. and Audouin et al. made at the n\_TOF facility have been finalized.

•Both sets of measurements tend to support the Lisowski *et al.* data somewhat better than the Shcherbakov *et al.* data. Lisowski has been investigating the hydrogen data used in converting his various ratio measurements to absolute cross sections. This work is on-going.

I recently received the data from Audouin et al. It is shown on the next slide.

**Final Audouin results compared with other measurements and the standards evaluation** 



NGT National Institute of Standards and Technology • Technology Administration • U.S. Department of Commerce

## <sup>235,238</sup>U(n,f) and <sup>239</sup>Pu(n,f) Measurements (cont.)

As noted at the last CSEWG meeting, there is still concern about the <sup>239</sup>Pu(n,f) cross section. The most recent data, by Tovesson and Hill agree reasonably well with the ENDF/B-VII standards evaluation and the Lisowski et al. and Shcherbakov et al. measurements up to about 10 MeV. The new measurements have somewhat smaller uncertainties than these other two data Sets. Above 10 MeV these new measurements fall somewhat lower than the ENDF/B-VII evaluation and the Lisowski et al. and Shcherbakov et al. measurements fall somewhat lower than the ENDF/B-VII evaluation and the Lisowski et al. and Shcherbakov et al. measurements except above about 100 MeV where they agree with the Lisowski et al. data.

•Additional work on actinide cross sections in the MeV energy is expected from a collaboration initiated by staff at LANL, LLNL and INL with several universities.

•This work will use Time Projection Chambers for fission detection. Very accurate measurements are anticipated with these detectors.

### **Additional Standards Work**

•In order to improve the standards on a continuing basis, an IAEA Nuclear Data Development Project "Maintenance of the Neutron Cross Section Standards" was initiated.

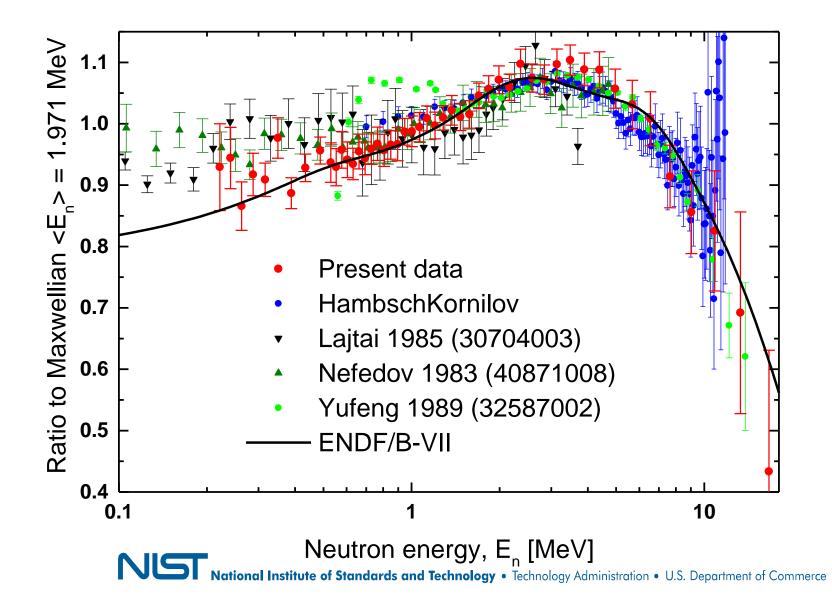
•This project has pursued improvements in the experimental database, considered additional standards, maintained evaluation codes and will periodically update the standards so they are available for new versions of data libraries.

### **Recent Activities Related to the Data Development Project (cont.)**

#### •Neutron spectra

• No new measurements have been made of the  ${}^{252}$ Cf spontaneous fission neutron spectrum. There are new measurements of the  ${}^{235}$ U(n<sub>th</sub>,f) neutron spectrum made by Kornilov (Hambsch) et al. and Vorobyev et al.

<sup>235</sup>U(n<sub>th</sub>,f) neutron spectra, "Present data" is Vorobyev et al.

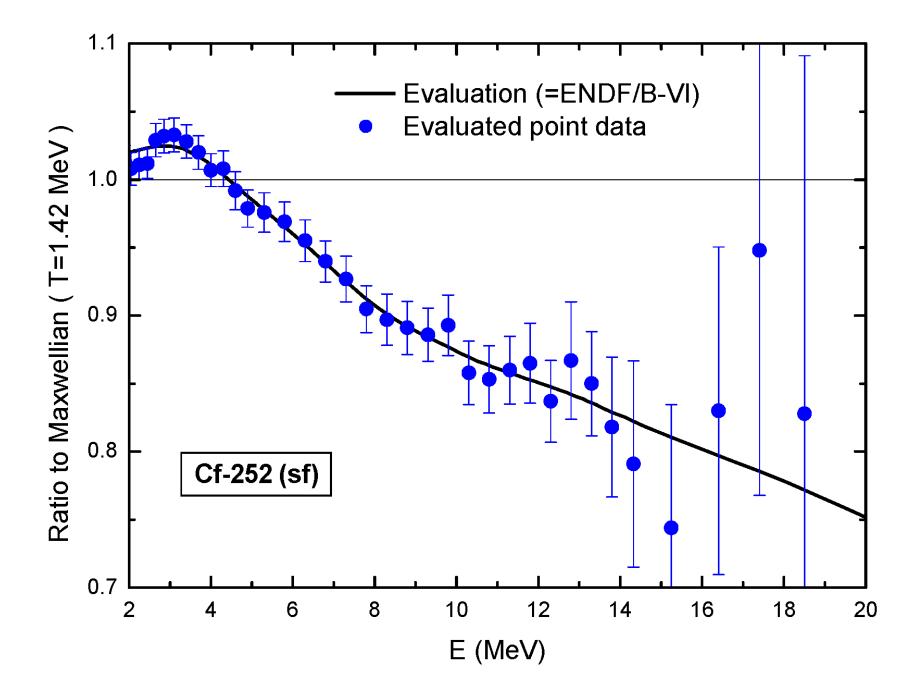


### **Recent Activities Related to the Data Development Project (cont.)**

•Neutron spectra (cont.)

• The most recent measurements of the  ${}^{235}U(n_{th},f)$  neutron spectrum have been made with a  ${}^{252}Cf$  source located outside the beam. Thus ratio measurements of these spectra were obtained.

• The GMA code used in the standards evaluation that can properly evaluate ratio data is being used to simultaneously evaluate these two fission spectra. Then there is an impact on both quantities in the ratio. This was done and it included smoothing using a model. Work continues to improve the results. This is an ongoing project.



### **Recent Activities Related to the Data Development Project (cont.)**

•Reference cross sections for measurements of prompt gamma-ray production cross sections. (cont.)

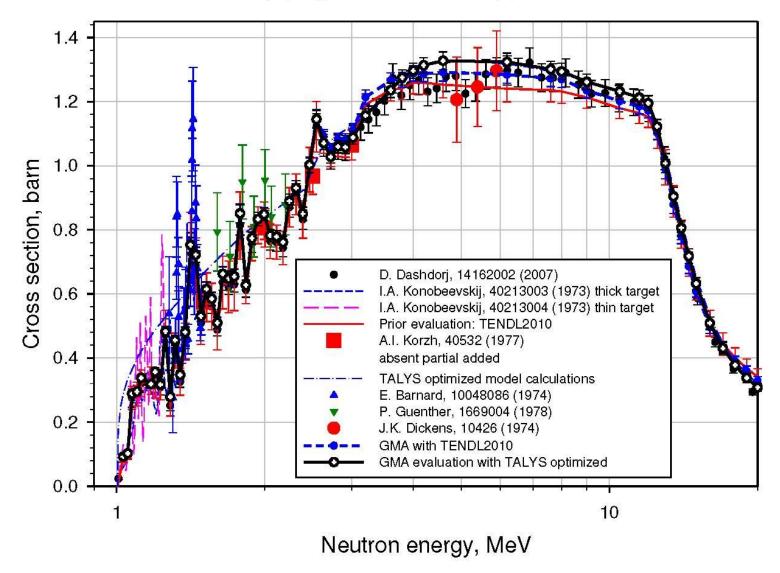
•Many nuclides and reactions were considered

• <sup>nat</sup>Ti with large yields of two gamma-lines, 984 keV from <sup>48</sup>Ti(n,n' $\gamma$ ) and 160 keV from <sup>48</sup>Ti(n,2n $\gamma$ ) and <sup>47</sup>Ti(n,n' $\gamma$ ) reactions appears to be one of the most suitable for use as a reference cross section. More work needs to be done to improve the experimental database.

•New measurements by Nelson using GEANIE have been made and are being analyzed.

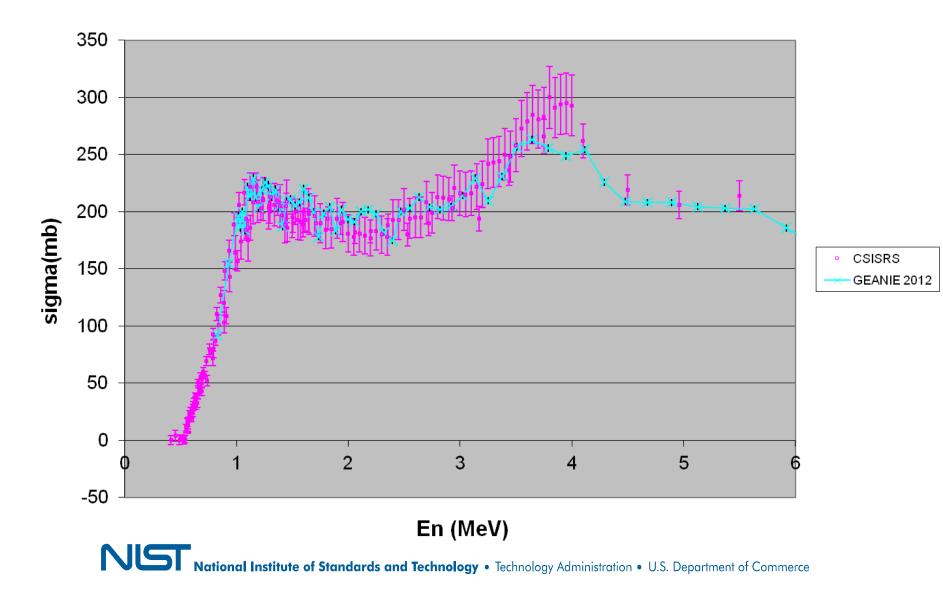
- •An improved evaluation by Simakov has been done.
- $Li(n,n'\gamma)$  also appears to be a reasonable candidate
  - •New measurements have been made by Nelson with GEANIE
  - •There is little high quality data at higher neutron energies except the Nelson work

# <sup>48</sup>Ti(n,n' $\gamma$ ) 983.539 keV $\gamma$ -production



NGT National Institute of Standards and Technology • Technology Administration • U.S. Department of Commerce

Li(n,n'gamma) Preliminary Results - Normalized to Previous Data



## Conclusions

•Recent experimental activity has improved the quality of the standards database. In most cases the data are in reasonable agreement with the evaluation. Areas of concern are:

•H(n,n) at small angles in the CMS near 15 MeV

•H(n,n) at intermediate and high energies where data are sparse and typically not available for a large angular range.

•Both <sup>6</sup>Li(n,t) and the <sup>10</sup>B standards need additional work as the emphasis is on extending the energy range to higher energies

•Additional work should be done in the high energy region on the  $^{235}$ U(n,f),  $^{238}$ U(n,f) and  $^{239}$ Pu(n,f) cross sections to support of the needs for better standards in that energy region .

•More work should be done on prompt gamma-ray reference cross sections and the gold capture cross section at low energies.

• The standards should be at the forefront, producing high accuracy cross sections including energy regions that may shortly require improved standards. It is short sighted to not have quality standards whenever they may be needed.