^{181}Ta - fast neutron evaluation

Profusely illustrated

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ENDF/B-VIII.0 evaluation

- Originally ENDL-2009 evaluation created by running Talys-1.0 with default input parameters. In this context, ENDF/B-VIII.0 performs quite well although we will show a number of deficiencies.

- Some features:
  - spherical Koning-DeLaroche potential most likely with the CC,
  - Hauser-Feshbach with Moldauer width fluctuation and ang. distributions,
  - Gilbert-Cameron level densities,
  - two-gas exciton model for the pre-equilibrium emission,
  - isomeric cross sections (MF=10) not included.

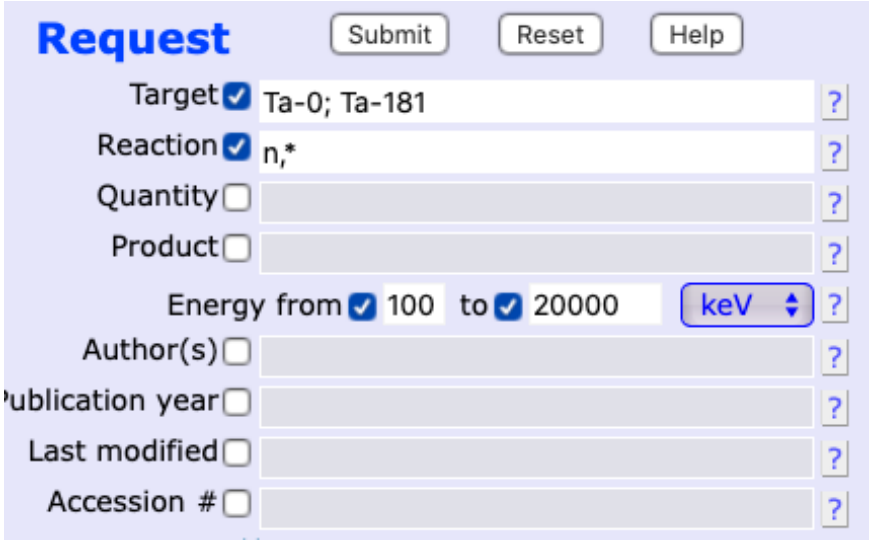
… a bit of motivation

- ENDF/B_VIII.0 description: “data file …is part of a large collection of isotopic evaluations…not tested against experimental data ... only as good as the global quality of Talys. ”

- Tantalum is used for casting of plutonium peats at LANL PF-4. k_eff for this casting is 2-3 times more sensitive to inelastic, elastic and capture on Ta than to the respective reactions on Pu239 (LA-UR-19-31175)
- Isomers abound in residual nuclei produced in interaction of fast neutrons with Ta so it is relevant to Radchem (LA-UR-21-23034)
- Evaluation of the resolved and unresolved region by ORNL/NNL has been recently submitted to NNDC.

Experimental data for fast neutrons (100 keV - 20 MeV)

- Exceptional wealth of exp. data for practically mono-isotopic (99.99%) Ta181
  - cross sections:
    - total
    - elastic
    - inelastic scattering
    - capture
    - (n,p)
    - (n,2n)
    - (n,α), (n,t), (n,^{3}He), (n,nα),
- Angular distributions
  - elastic
  - inelastic
- n and γ DE and DDX (+ p & α)
- Several isomeric cross sections

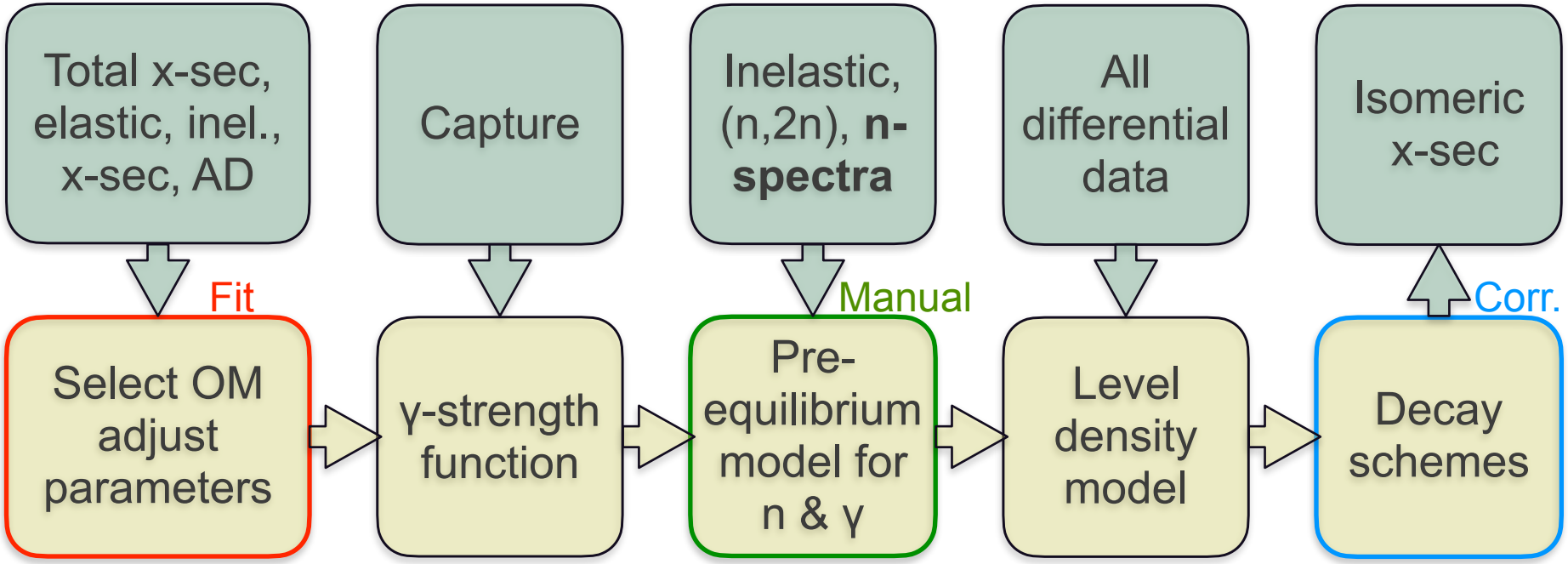


- Number of EXFOR datasets : 343
- Number of EXFOR quantities: 137
- EMPIRE makes 468 plots (!)

Extraordinarily well distributed measurements!

Concept of this evaluation

- Combine modeling of EMPIRE-3.2 with experimental data to produce a consistent picture of nuclear reactions considering “all” available differential and integral data.



Implications of the concept

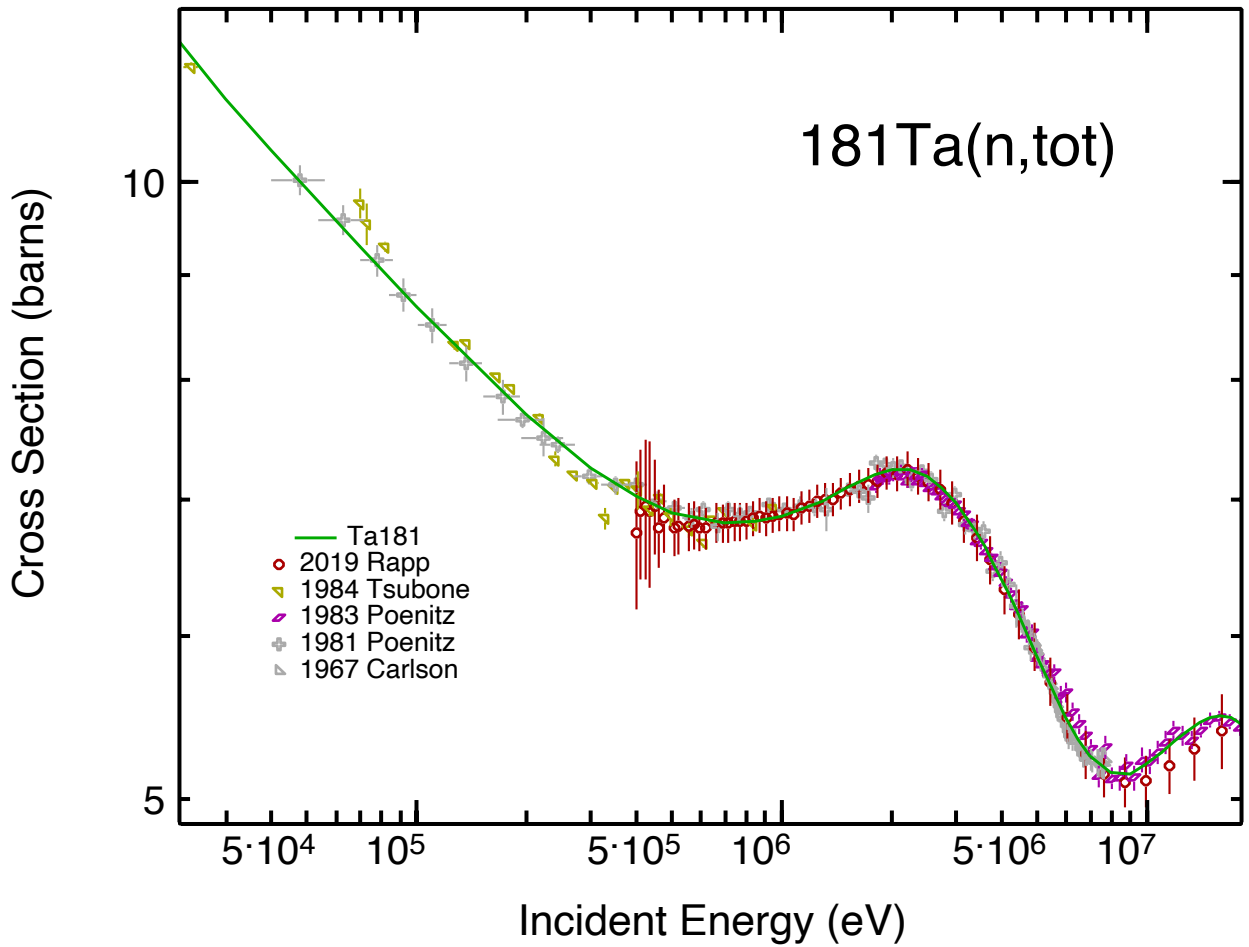
- Evaluation totally defined by the code and its input (physics preserved)
- Selection of models given priority over adjustment of parameters (right physics)
- Ignoring experiments inconsistent with other experimental data or with general picture
- **Gradual use of experimental data in the model selection and parameter adjustment** (“don’t fit OMP to capture”)
- Limited model parameter adjustment (mostly manual)
- Global fitting of model parameters only at the very end (…if needed)

All the results presented below were obtained without global parameter fitting!

CC optical model - total x-sec

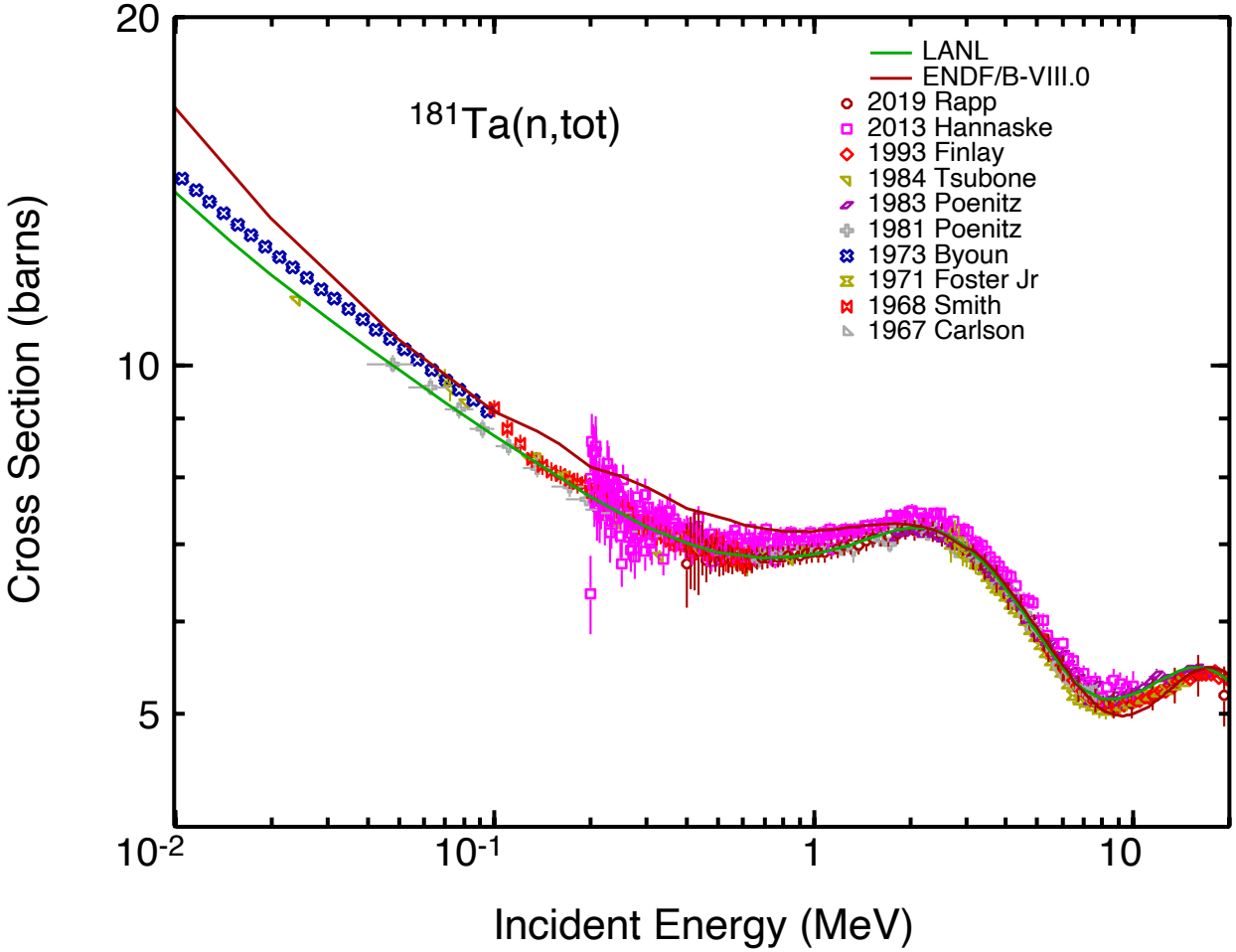
- Ta181 is a strongly deformed and requires Coupled-Channels (CC)
- The RIPL-3 (#610) regional CC potential extends up to 200 MeV but it needs adjustments from 50 keV up to 20 MeV
- Two reliable and consistent measurements of total by Poenitz et al, covering entirely this energy range were used to refine the potential
- Fitting performed with Kalman filter resulted in rather minor corrections

r_v	0.98542
V_v	1.0184
a_v	1.0870
β_2	1.0444
β_4	0.93707
β_6	0.49351



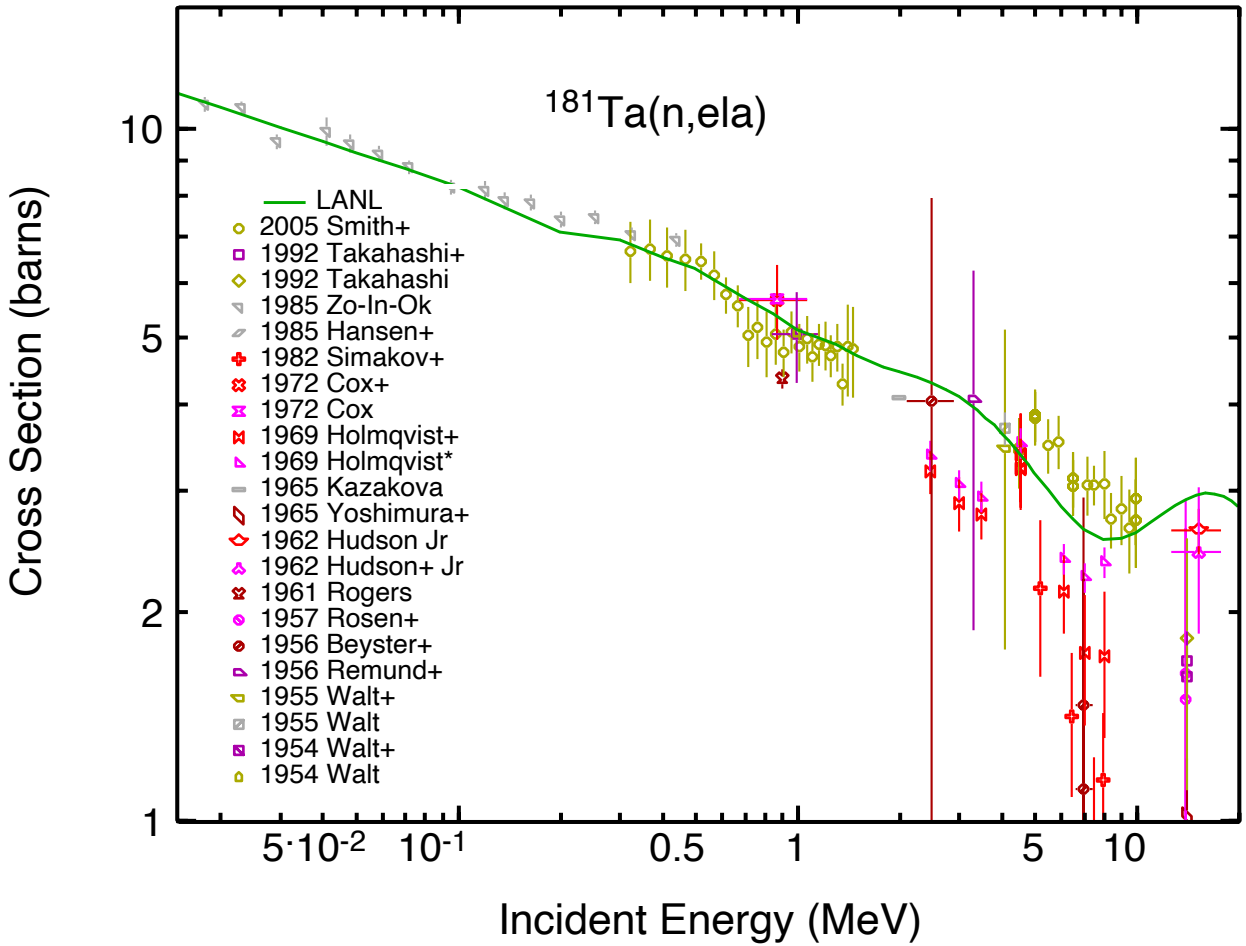
Five experiments that support the evaluated total compared with the CC calculations using updated optical potential.

LANL evaluation perfectly fits Rapp, Poenitz 81 & 83, Tsubone, Smith, and Carlson.



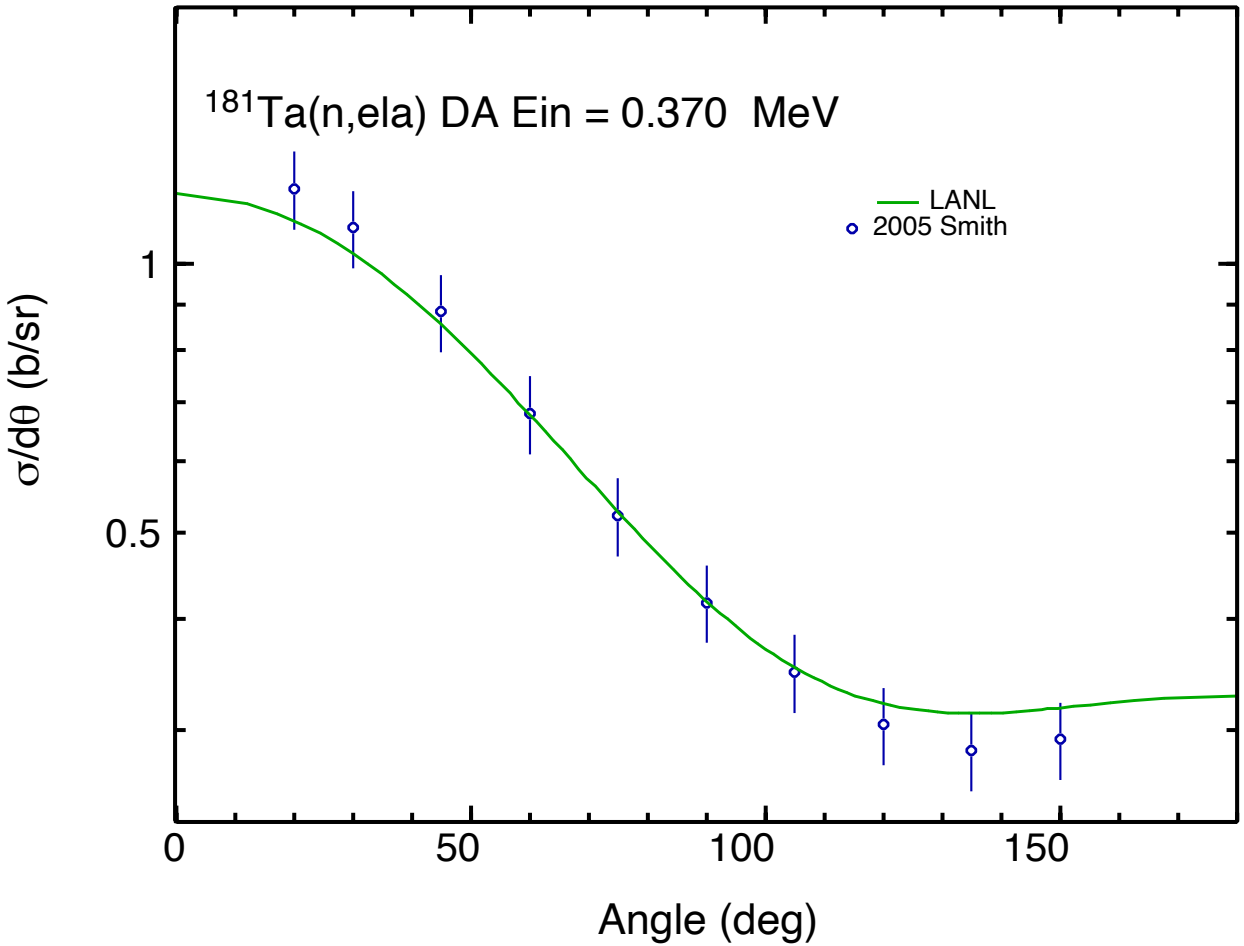
- Hannaske, and Byoun show the same shape as LANL evaluation and agree after ~3% and ~4.5% renormalization respectively. A 1.5% upscaling brings Foster into agreement.
- Finlay shape is slightly different but still within 2% of the new evaluation
- VIII.0 too high below 2 MeV and too low above 7 MeV

CC model - elastic x-sec & angular distributions

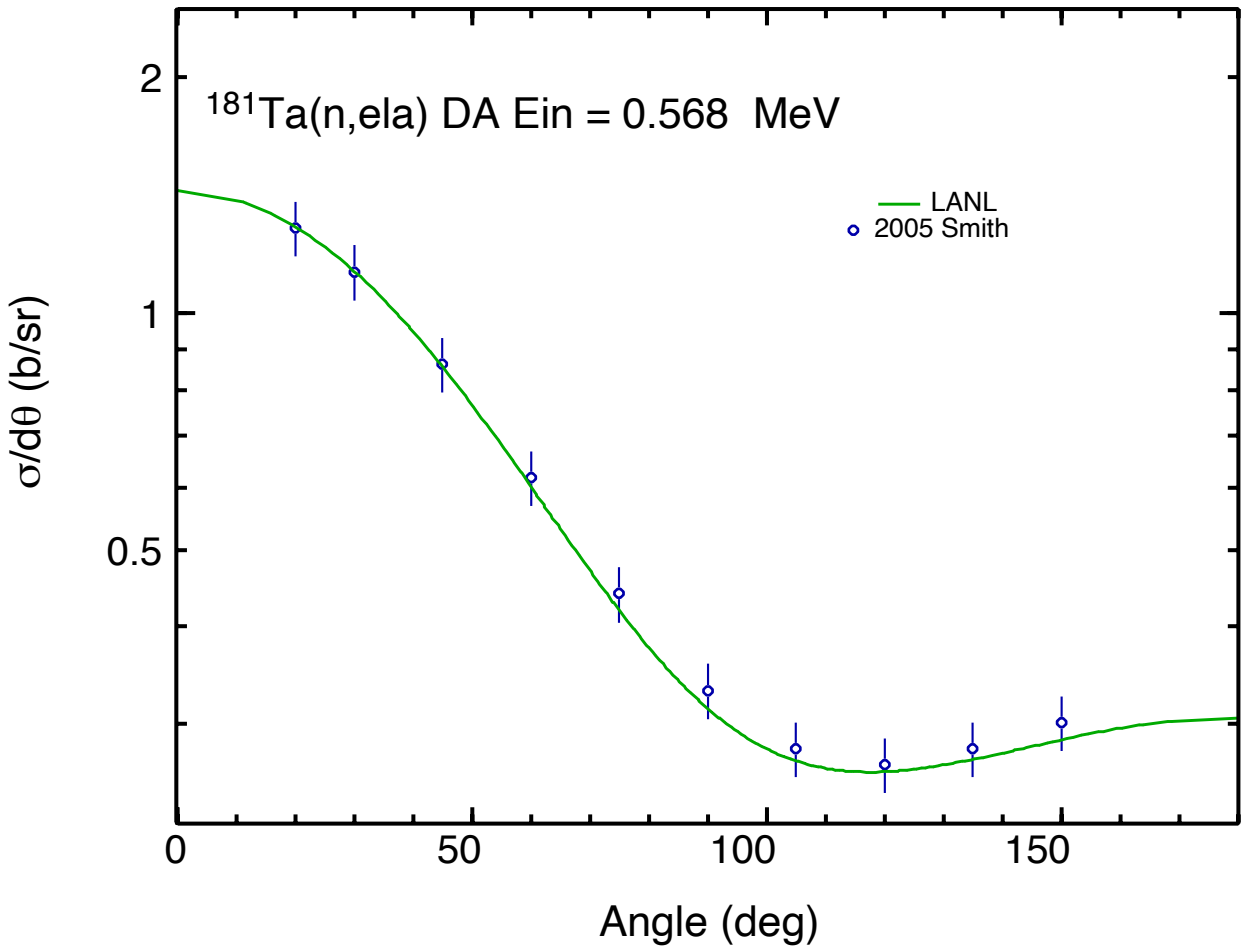


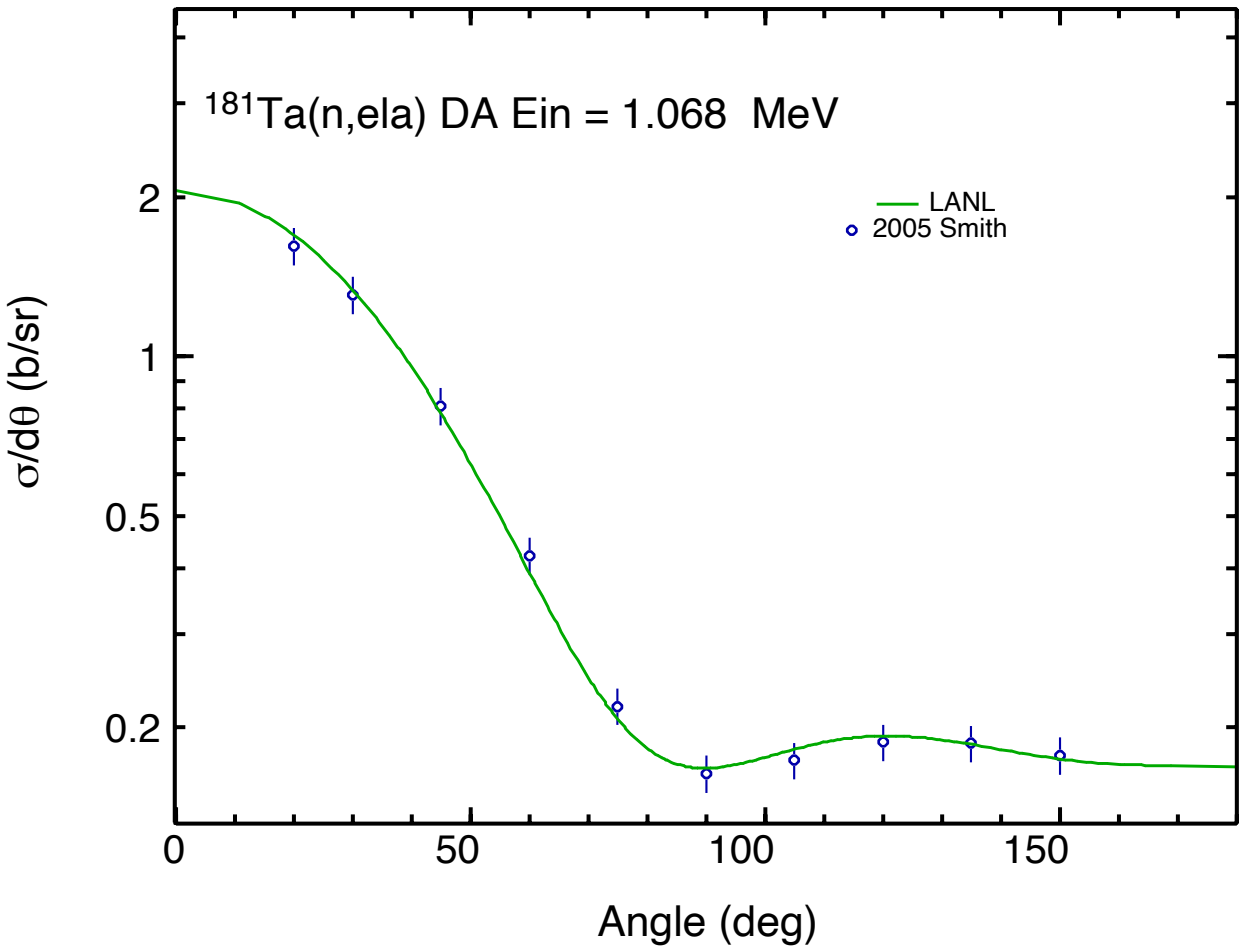
Elastic cross sections after adding the 1-st and the 2-nd inelastic.

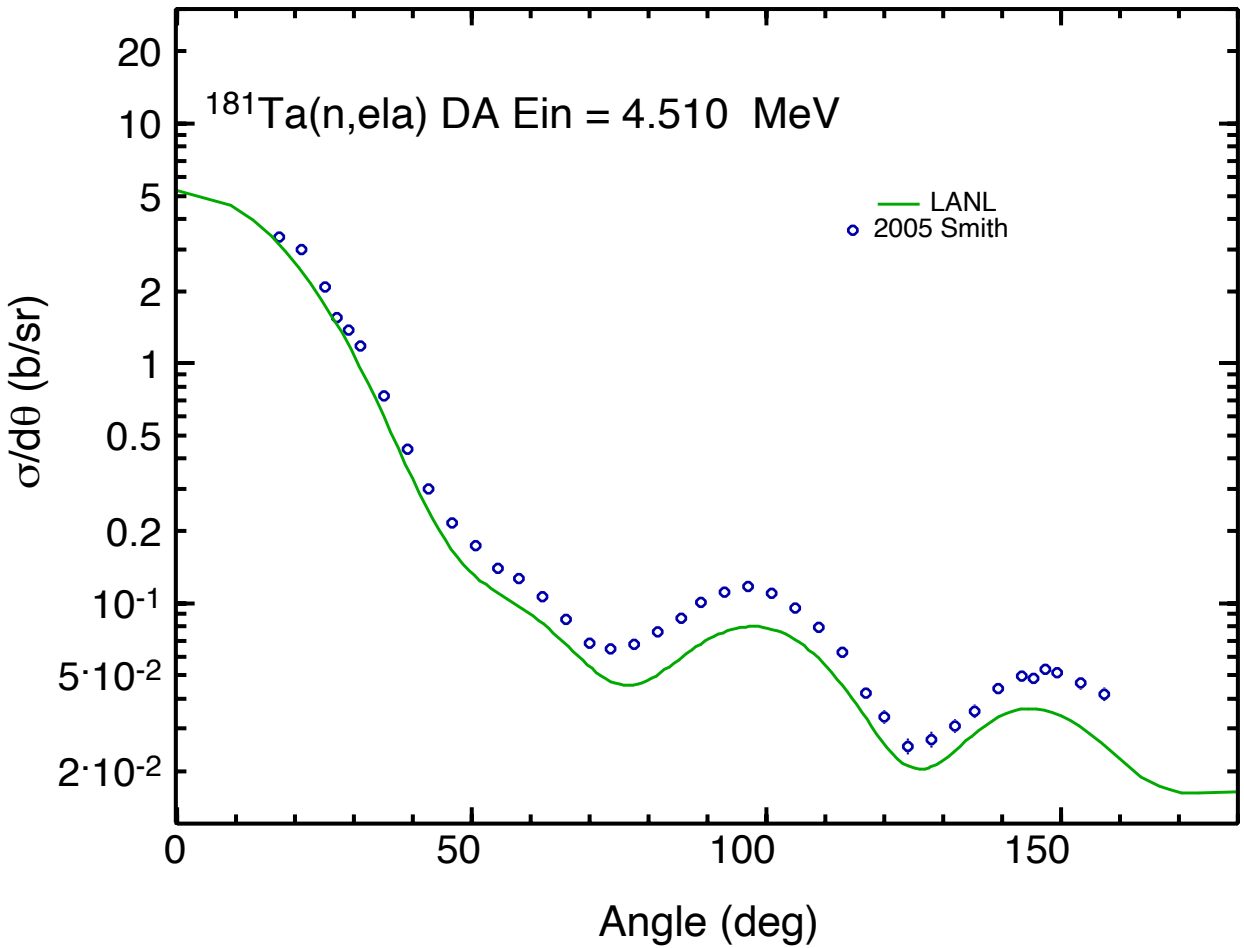
Elastic angular distributions



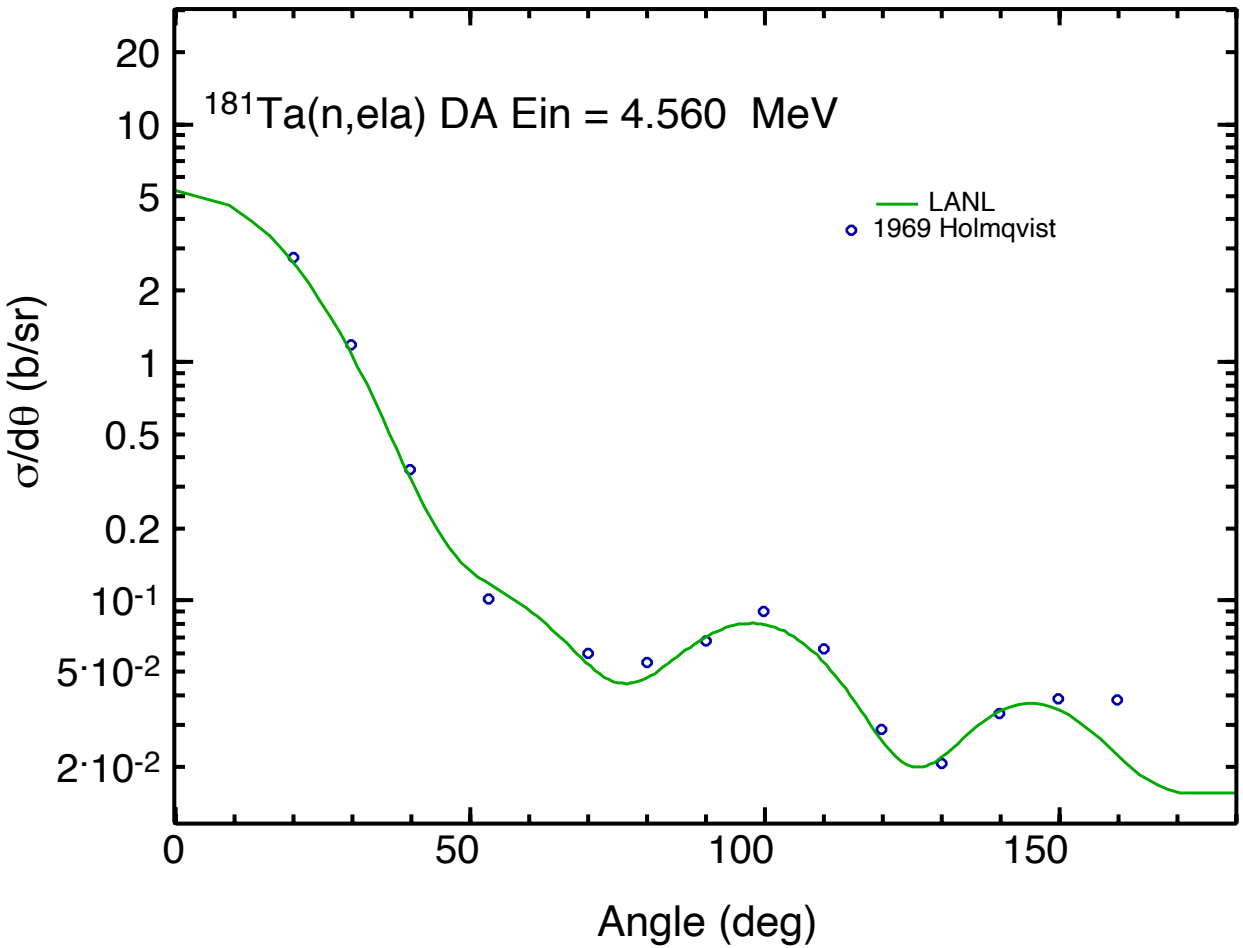
The 1-st and the 2-nd inelastic angular distributions added.


^{181}Ta(n,ela) DA Ein = 0.568 MeV
LANL
2005 Smith
σ/dθ (b/sr)
Angle (deg)
2
1
0.5
0
50
100
150

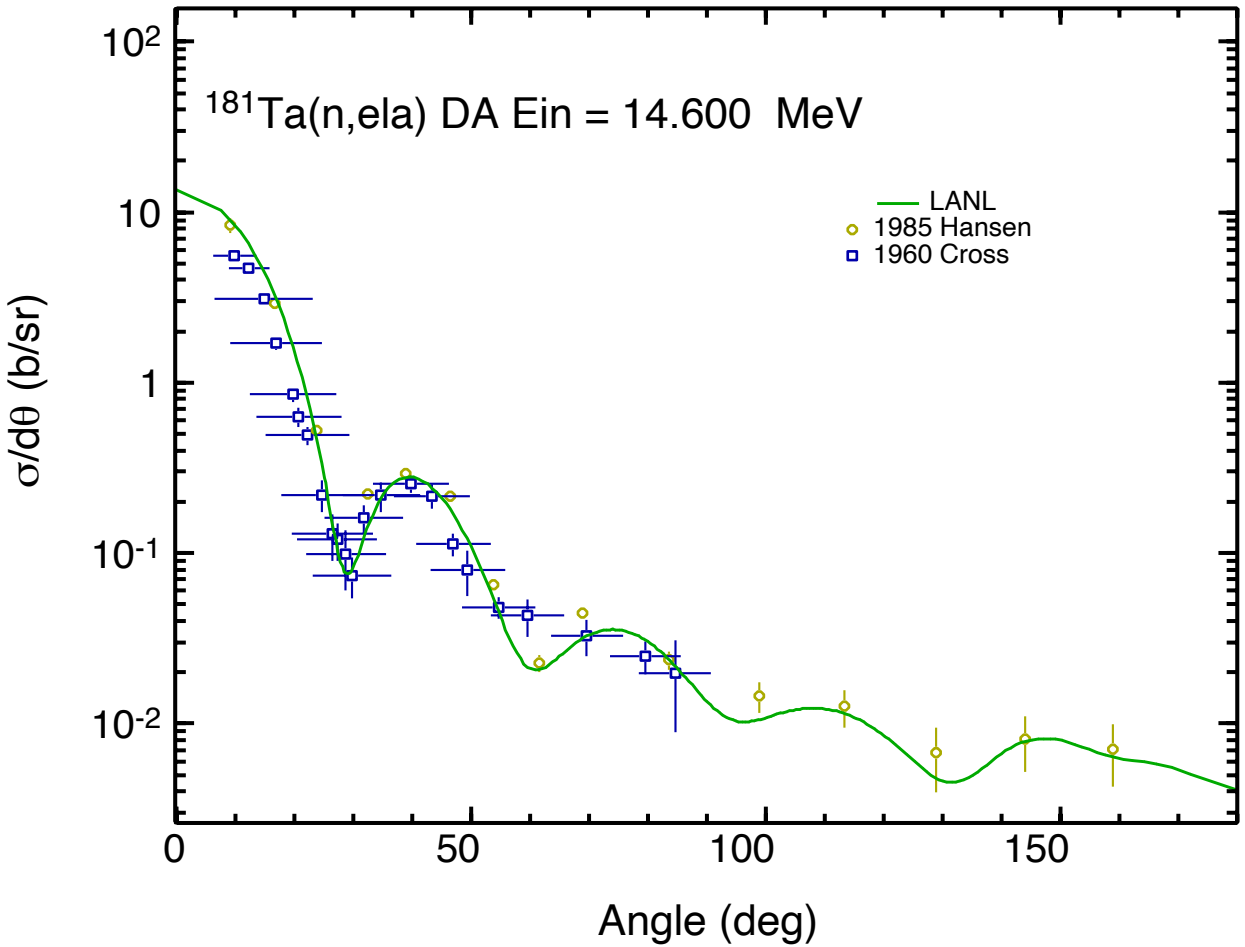

^{181}Ta(n,ela) DA Ein = 1.068 MeV
LANL
2005 Smith
σ/dθ (b/sr)
2
1
0.5
0.2
0
50
100
150
Angle (deg)



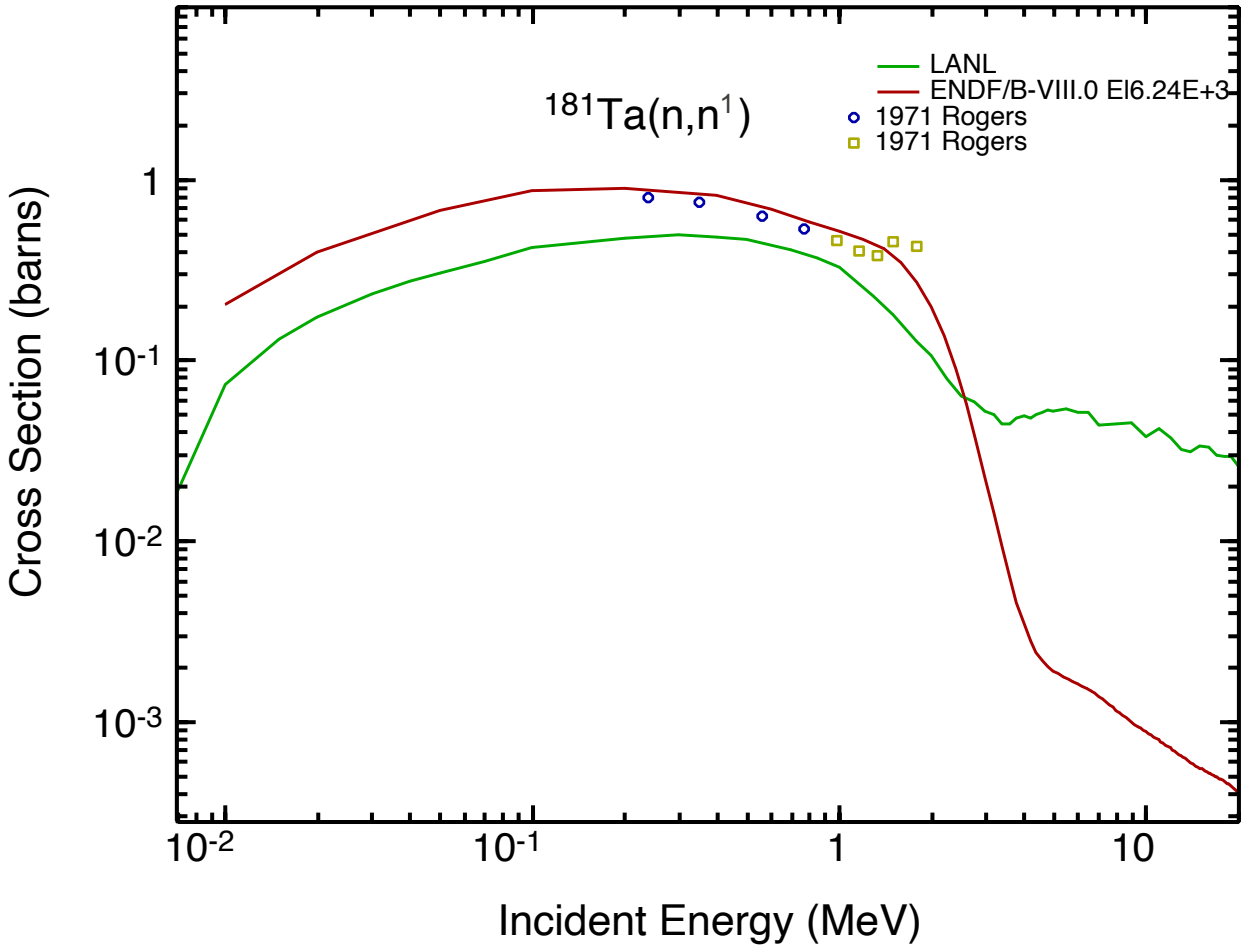
At 4.5 MeV calculations underestimate Smith data, although the shape is quite good. Other experiments, however, are also lower…



This trend continues through the higher incident energies - Smith data are higher than the evaluation while other experiments agree.


^{181}Ta(n,ela) DA Ein = 14.600 MeV
LANL
1985 Hansen
1960 Cross
σ/dθ (b/sr)
Angle (deg)
10^2
10
1
10^{-1}
10^{-2}
0
50
100
150

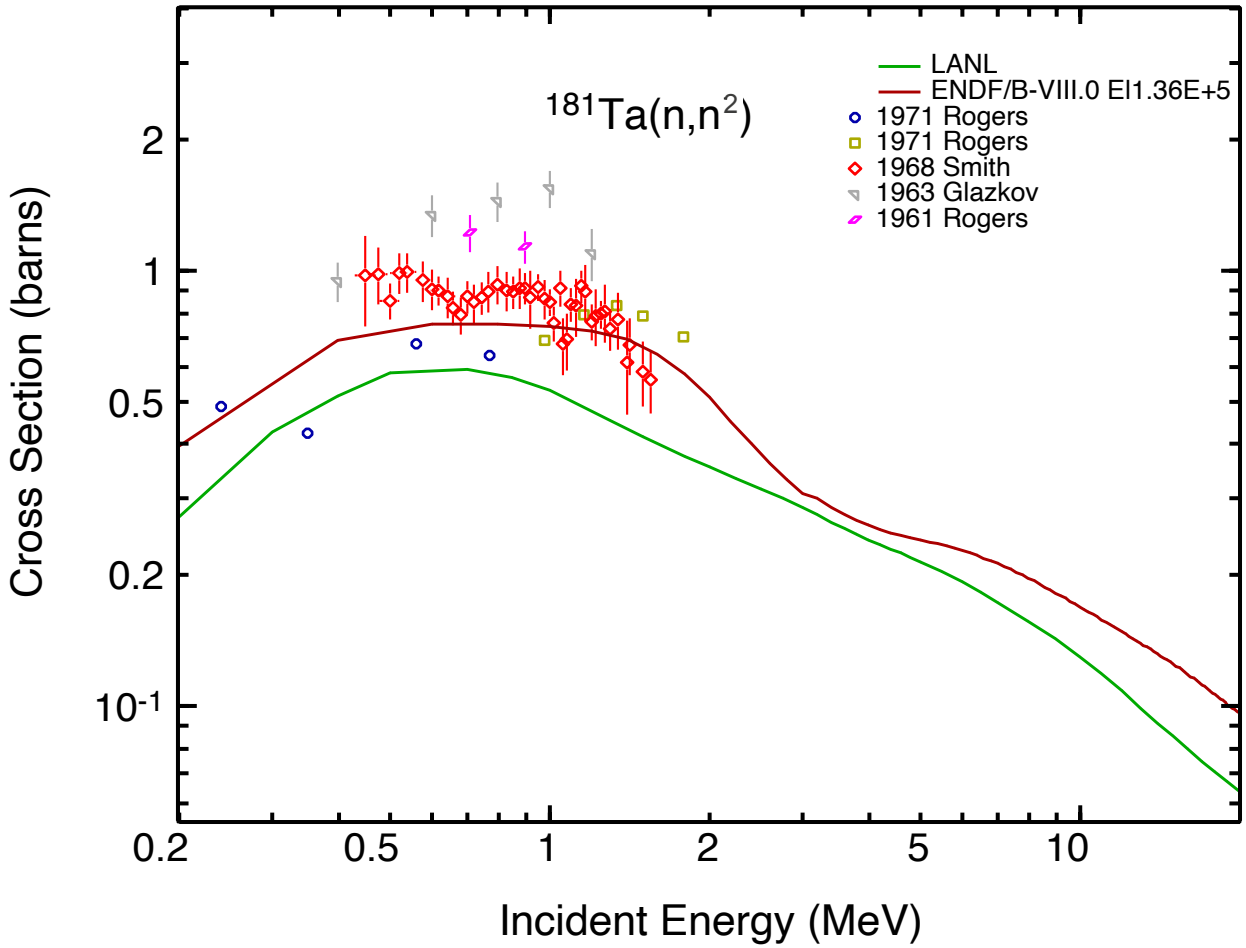
CC model - inelastic x-sec



The 1-st inelastic (summed into elastic) does not belong to the ground state rotational band. The DWBA has been added to increase elastic angular distr. at higher energies (disregard numerical fluctuations)

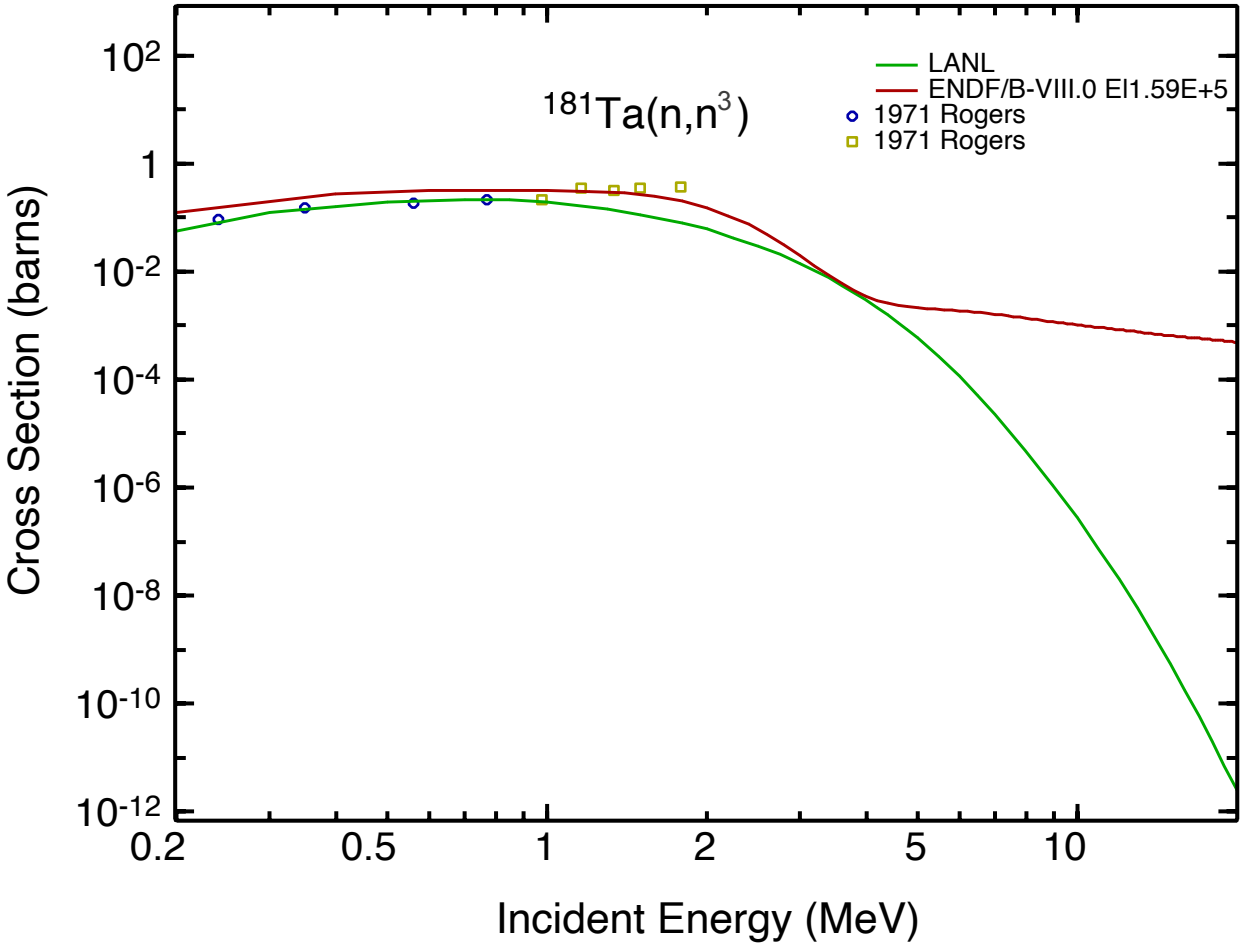
VIII.0 cross sections are higher for this and other inelastics and agree better with experiments but.. it comes at a price of overshooting total by ~0.5 b.

2-nd inelastic



The first collective level at 136 keV above the g.s. Similarly to the first it is higher in VIII.0.

3-rd inelastic



Not collective in LANL calculations. In VIII.0 seems to be collective but the high-energy tale might also come from the PE contribution distributed over all discrete levels.

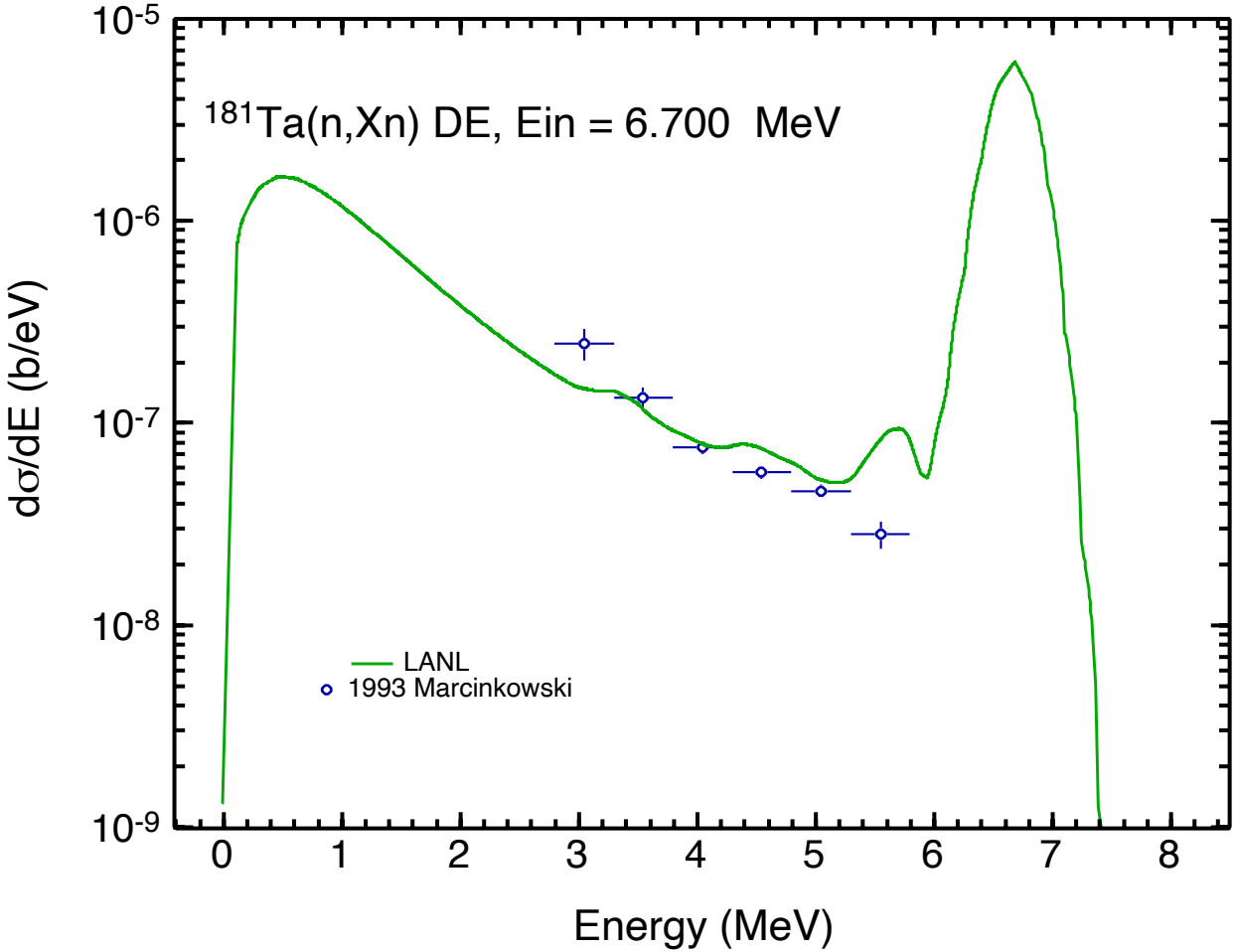
MSD/MSC - neutron spectra, inelastic & (n,xn)

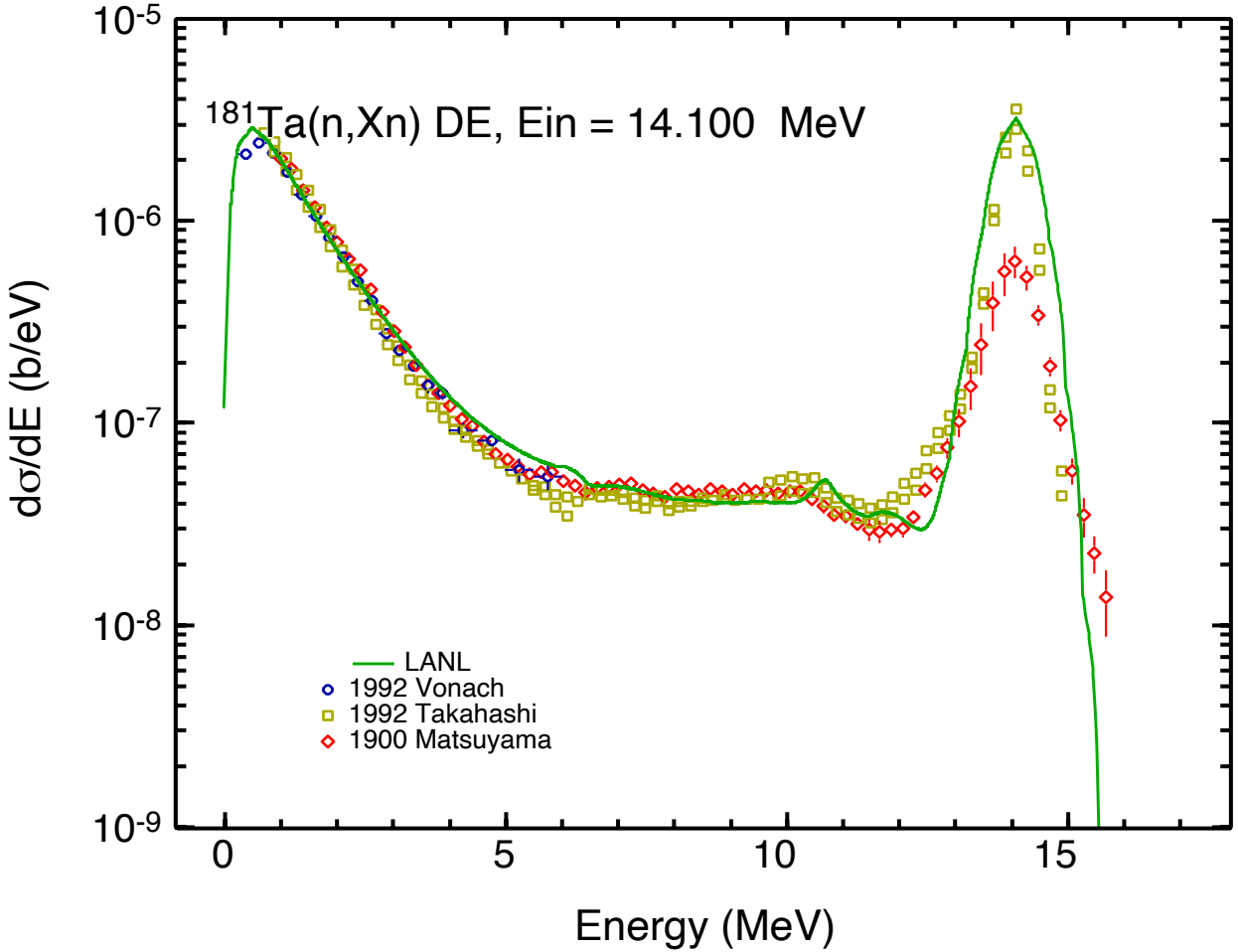
- Quantum mechanical Multistep Direct (MSD) and Multistep Compound (MSC) were employed for neutron emission instead of the exciton model.
- MSD eliminates the need of adding large number of collective discrete levels to the continuum. Default calculations were pretty good but adjusting (manually) a single parameter brought additional improvement.

MSD I_o = 6.38 MeV

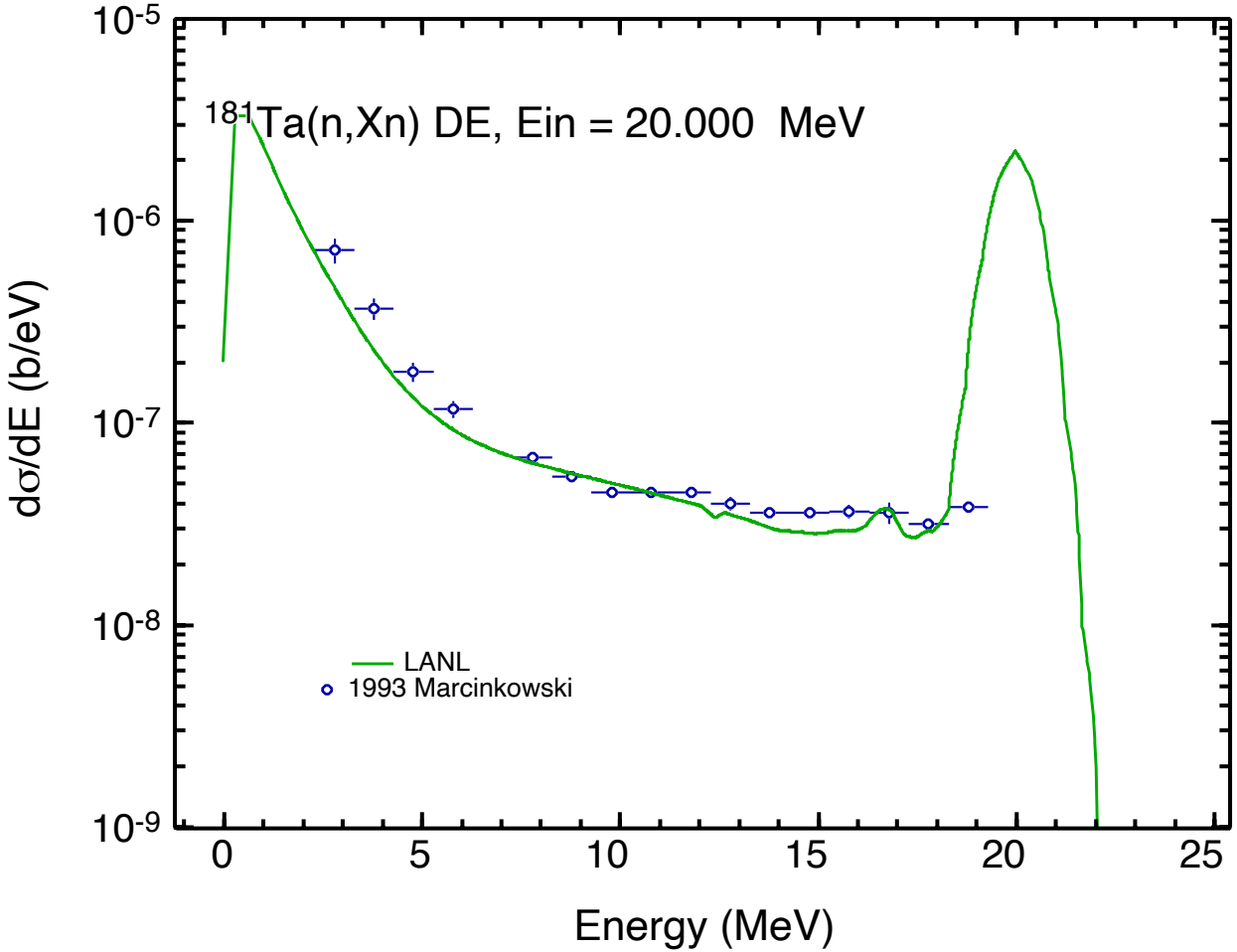
- Extensive experimental coverage of DDX from 5 to 20 MeV of incident energies at different angles was essential for building confidence in the calculations.

Angle integrated energy n-spectra

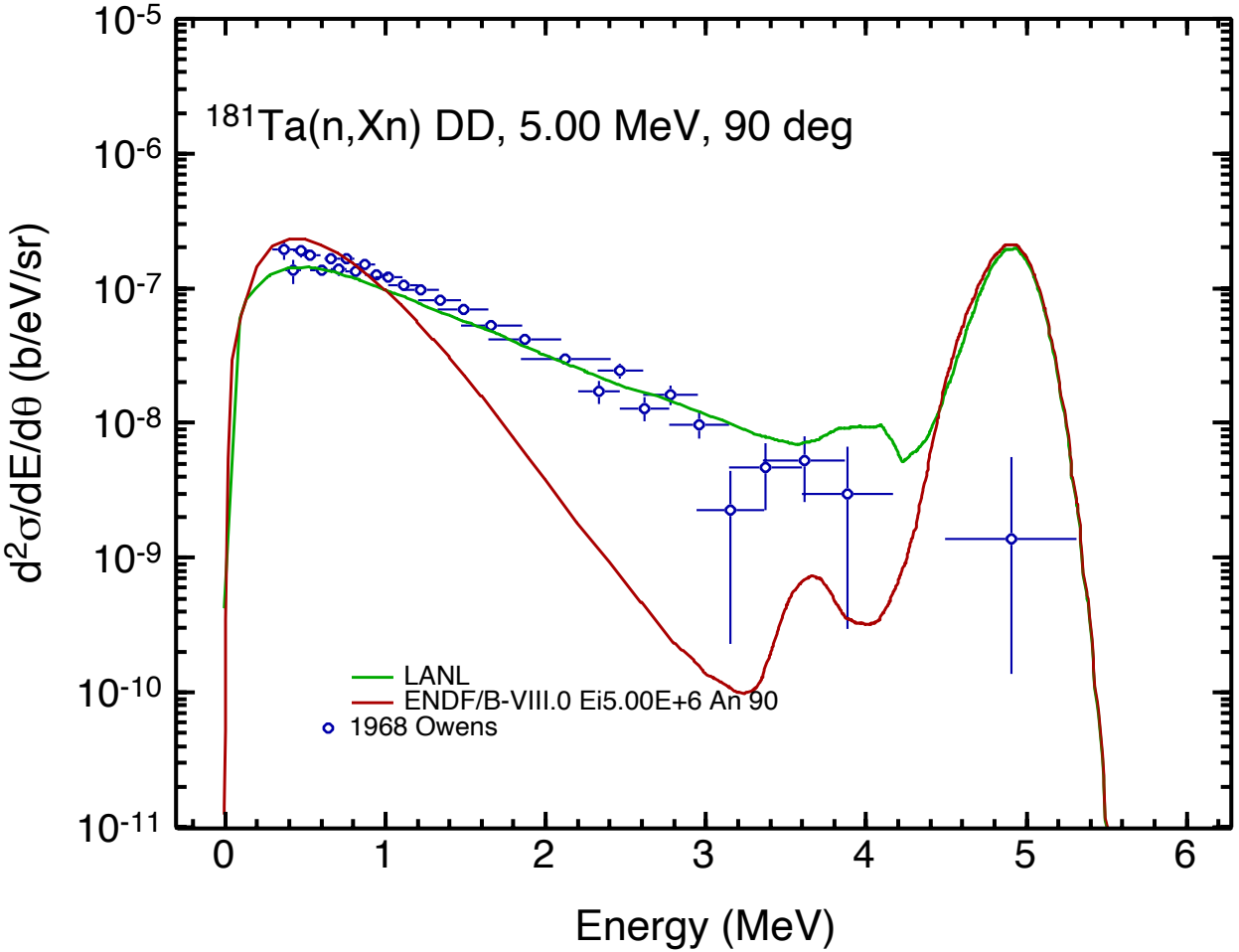




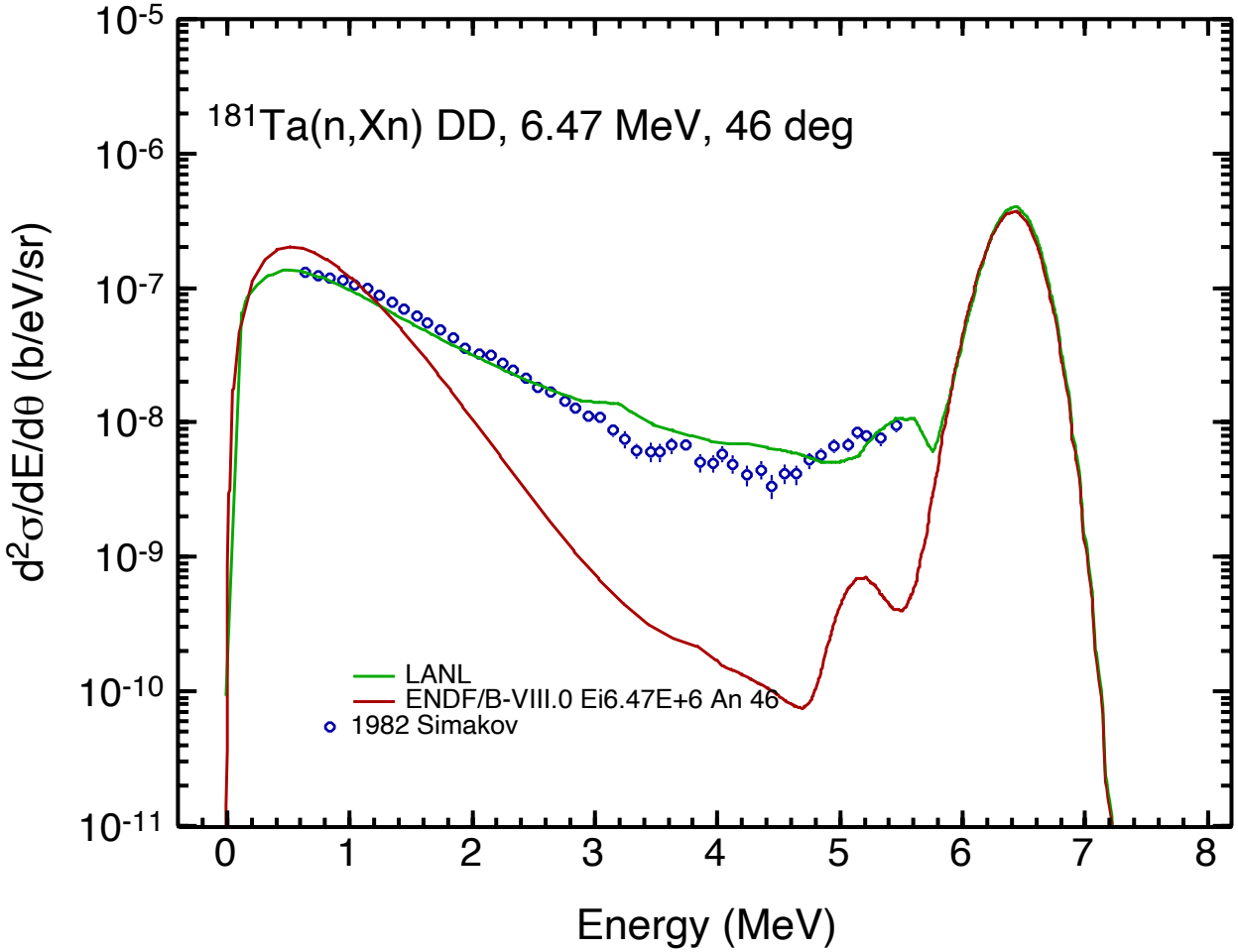
Three consistent data sets are well reproduced by the calculations.

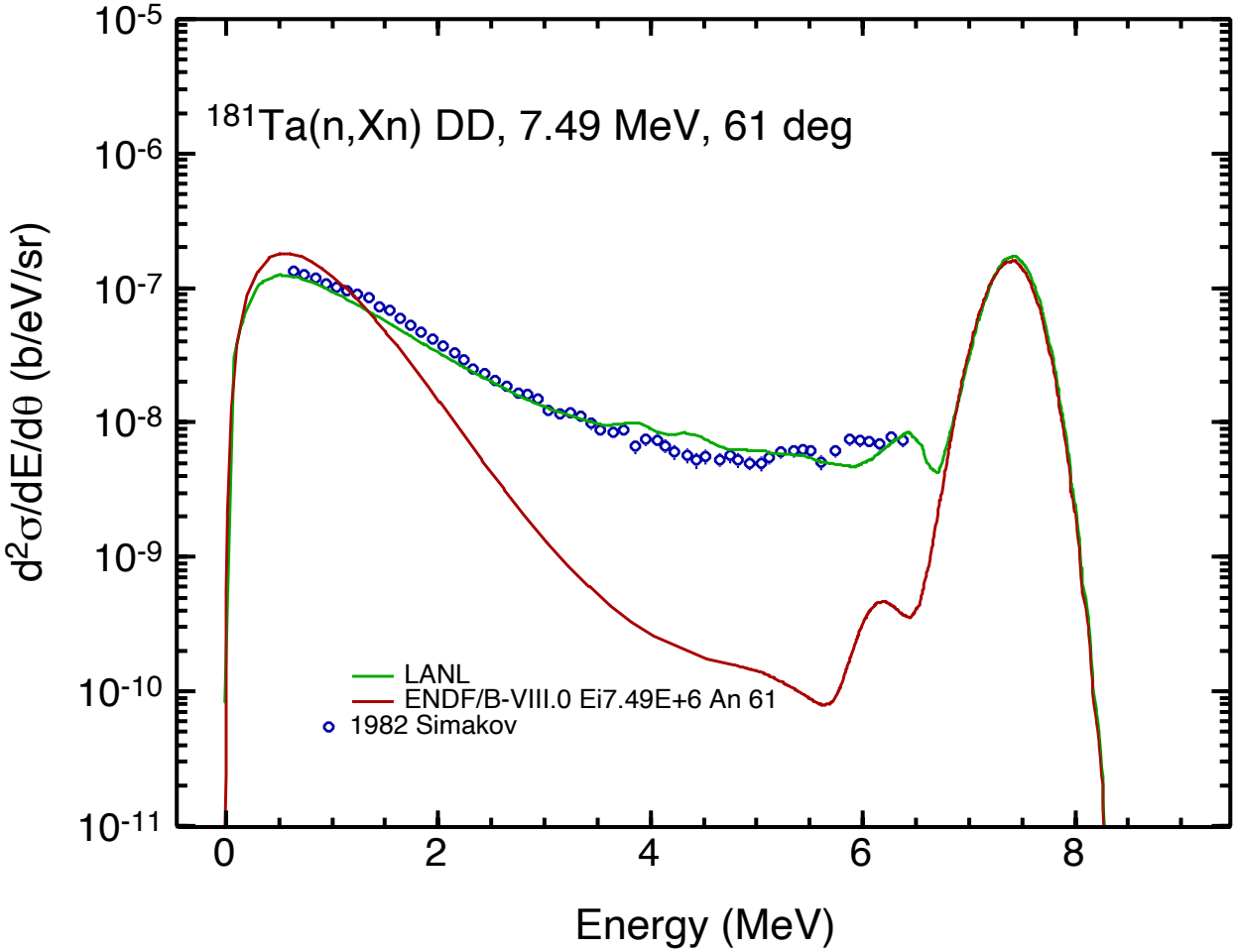

181Ta(n,Xn) DE, Ein = 20.000 MeV
dσ/dE (b/eV)
Energy (MeV)
LANL
1993 Marcinkowski
10-5
10-6
10-7
10-8
10-9
0
5
10
15
20
25

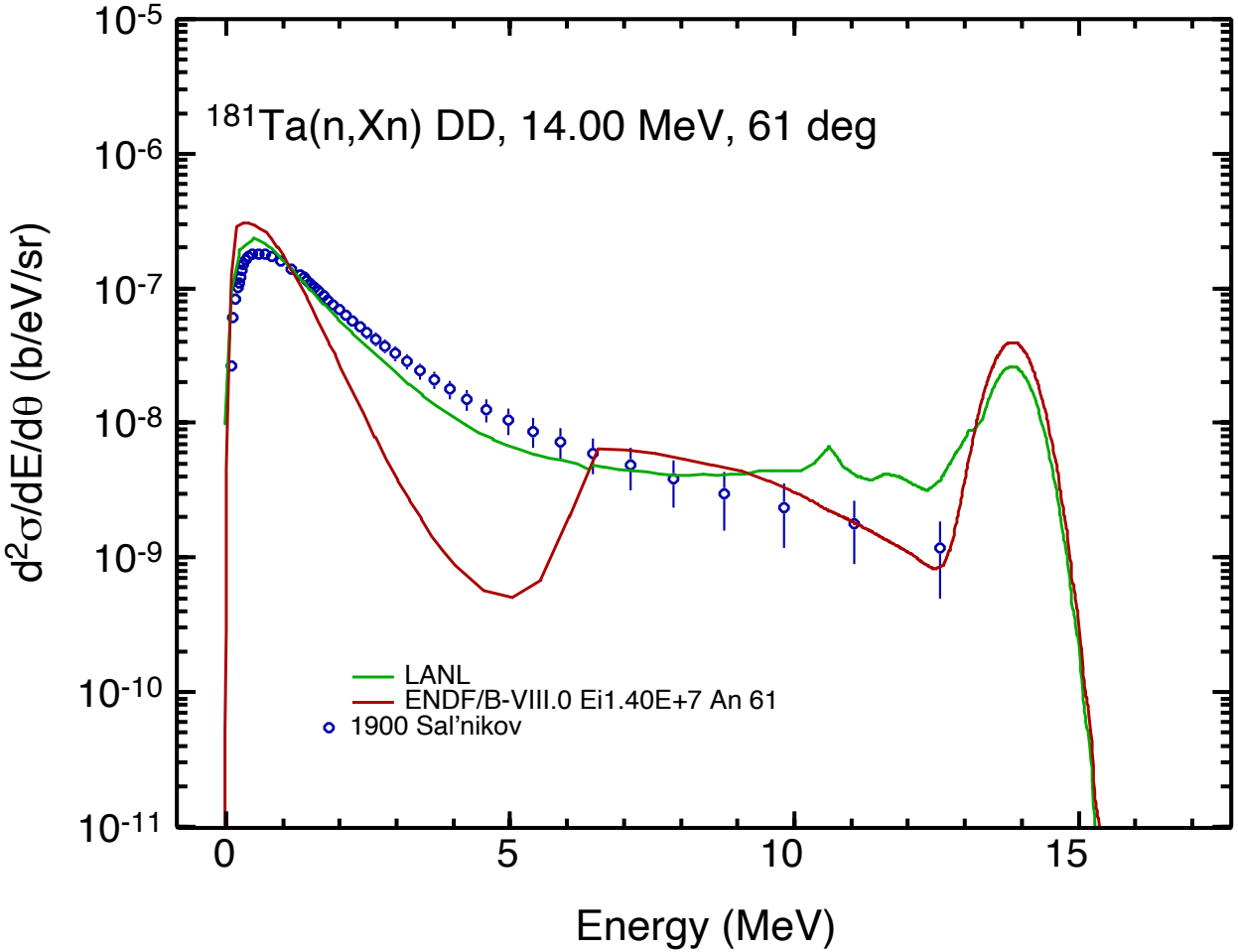
Double-differential n-spectra



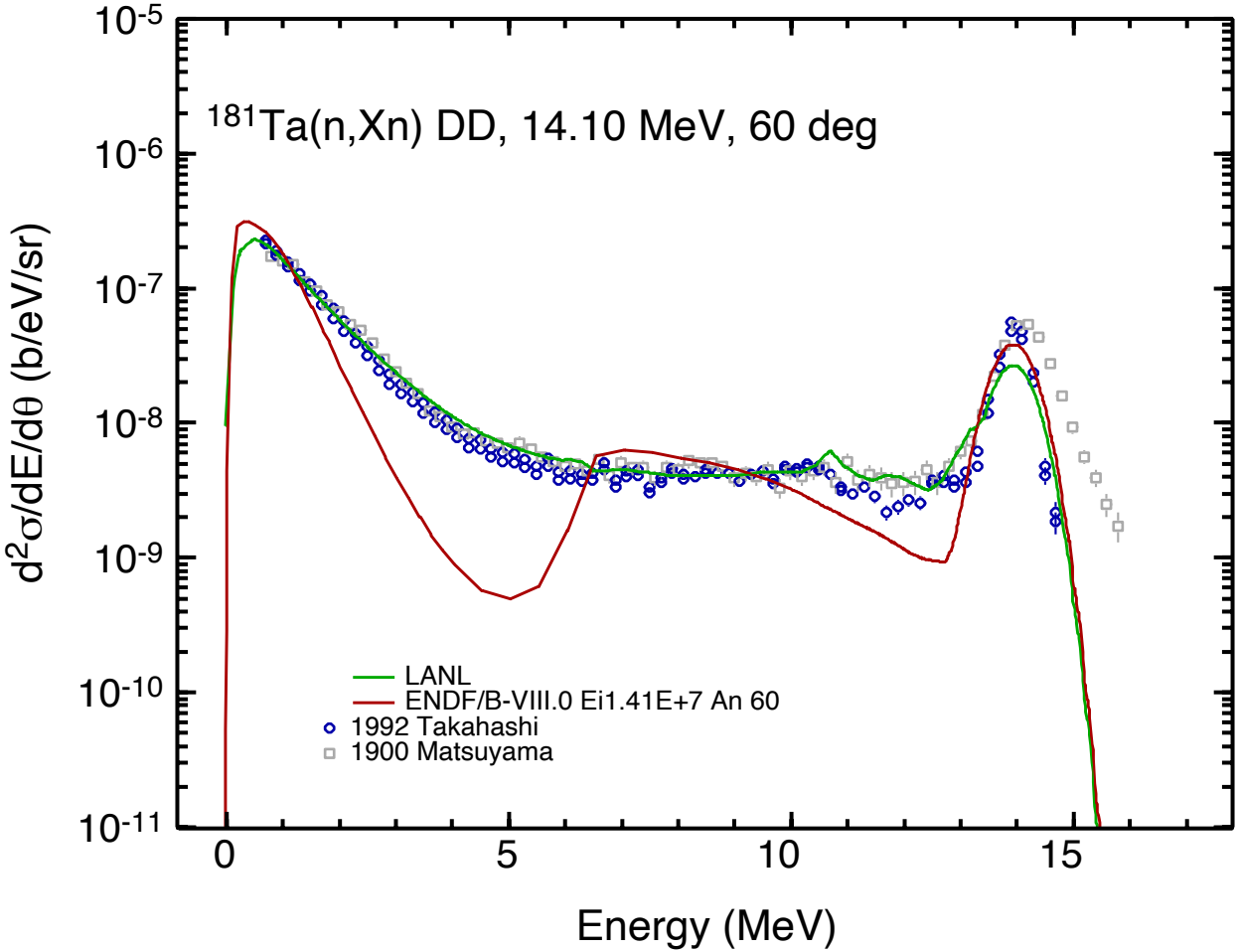
VIII.0 has a serious issue with all neutron spectra.


^{181}Ta(n,Xn) DD, 6.47 MeV, 46 deg
$d^2\sigma/dE/d\theta$ (b/eV/sr)
Energy (MeV)
LANL
ENDF/B-VIII.0 Ei6.47E+6 An 46
1982 Simakov
10^{-5}
10^{-6}
10^{-7}
10^{-8}
10^{-9}
10^{-10}
10^{-11}
0
1
2
3
4
5
6
7
8

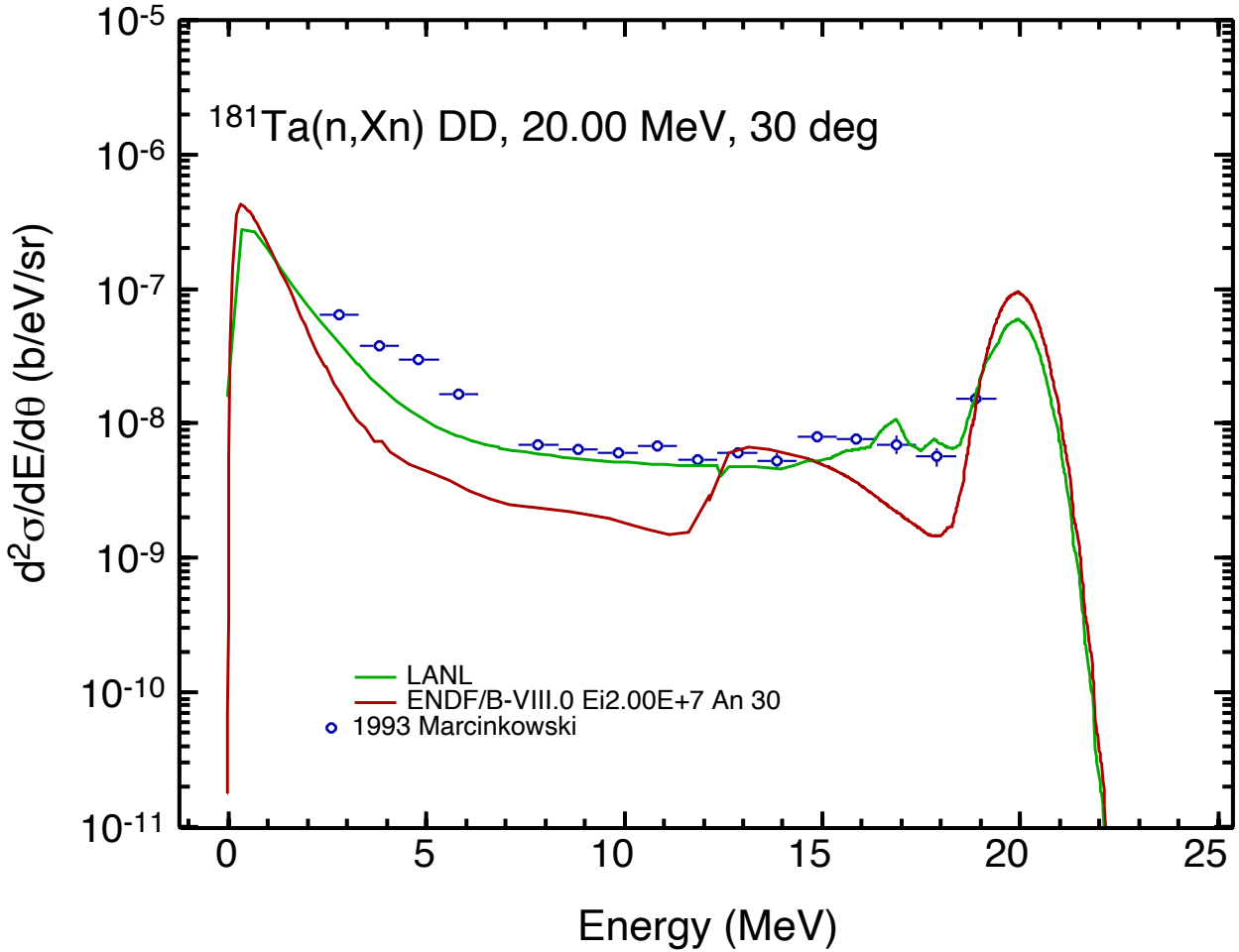

^{181}Ta(n,Xn) DD, 7.49 MeV, 61 deg
$d^2\sigma/dE/d\theta$ (b/eV/sr)
Energy (MeV)
LANL
ENDF/B-VIII.0 Ei7.49E+6 An 61
1982 Simakov
10^{-5}
10^{-6}
10^{-7}
10^{-8}
10^{-9}
10^{-10}
10^{-11}
0
2
4
6
8



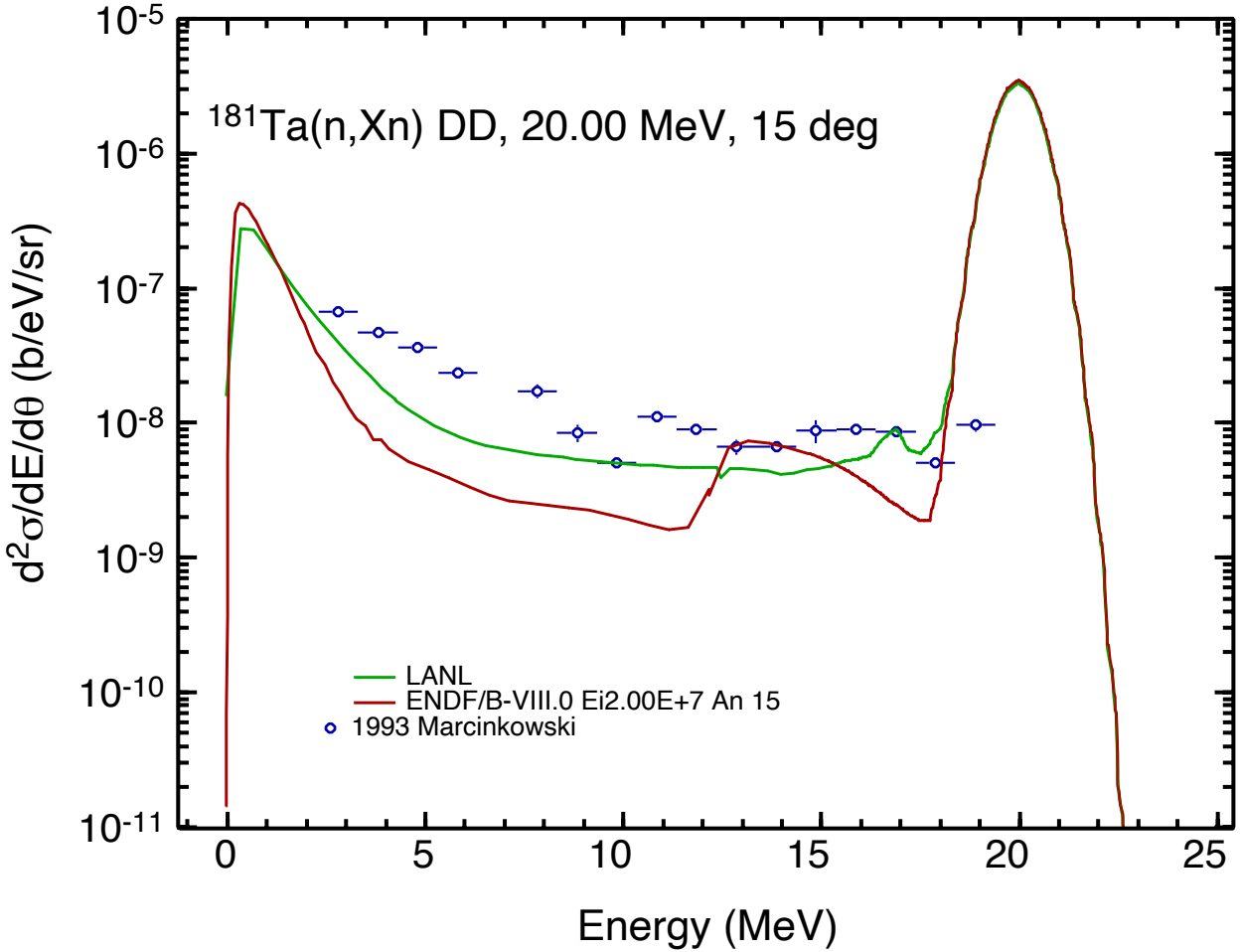
If Salnikov data were the only available it could be concluded that exciton model is enough to describe neutron spectra…



…which is obviously not true.



Even 30 deg at 20 MeV is well reproduced, but not 15!

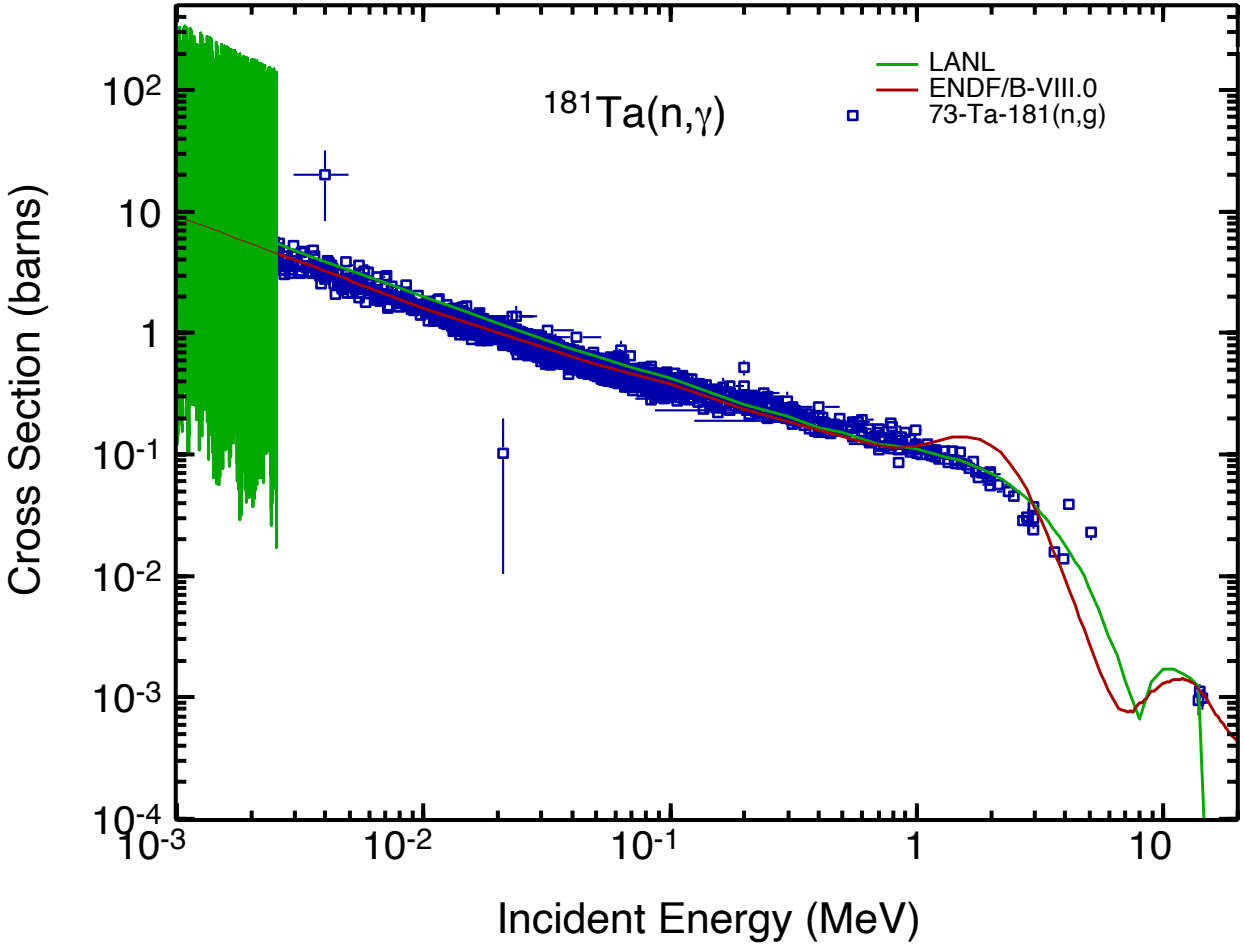


This is a recurring problem and I wonder whether it is related to experimental difficulties related to beam neutrons scattered to the detectors?

MSD could also be blamed (compressional form-factor overshoots forward angle data).

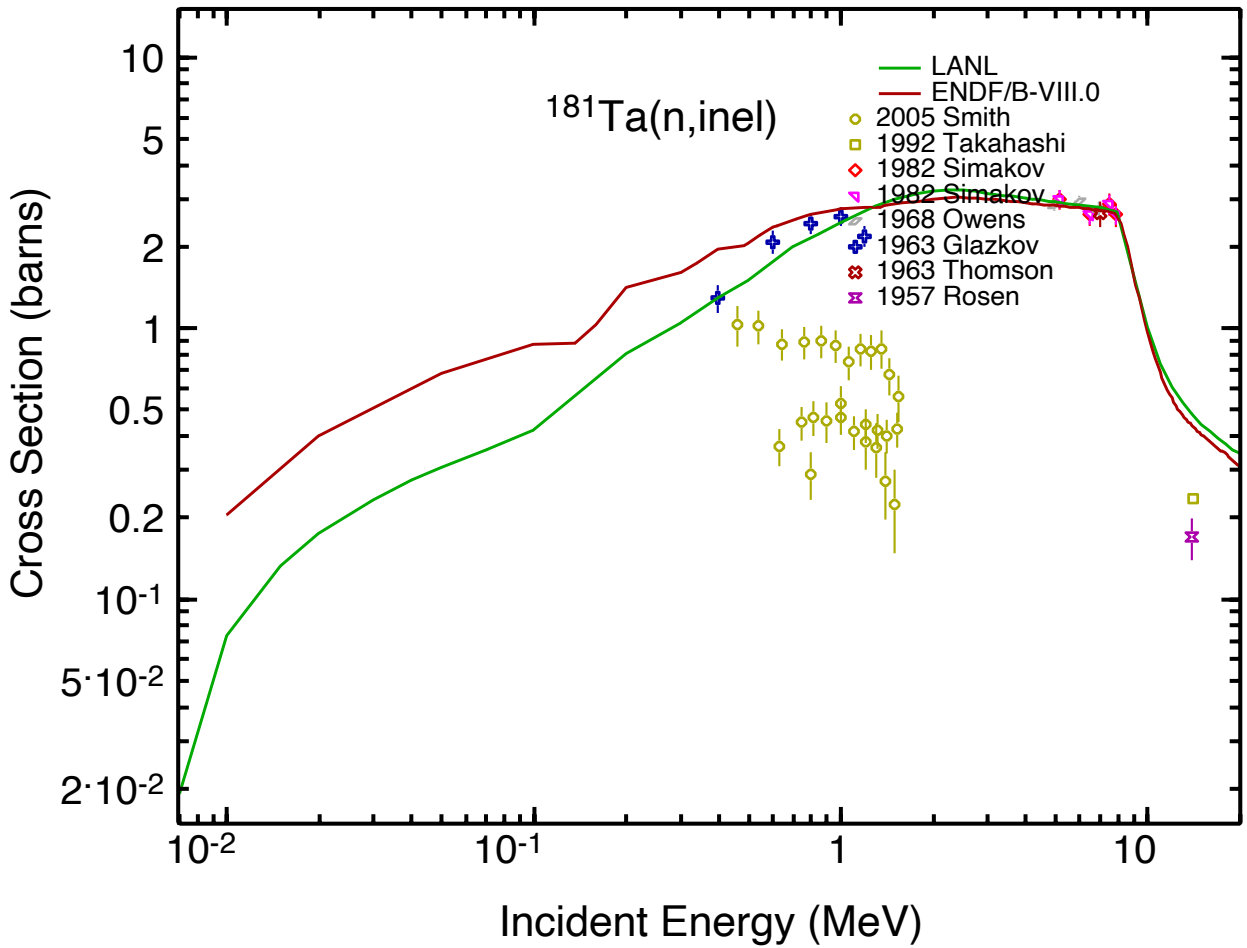
Here, however, we have dramatic increase of the data below 5 MeV that is hard to understand.

Level densities - capture, inelastic, (n,2n) x-sec, neutron & gamma spectra, …



- Gilbert-Cameron level densities were chosen over EGSM and microscopic Hartree-Fock-Bogoliubov because the former produce better capture cross sections between 1-3 MeV, and slightly better gamma spectra.
- The effect on the neutron spectra is mixed with GC working better at incident energies around 14 MeV while EGSM has an advantage at 20 MeV.

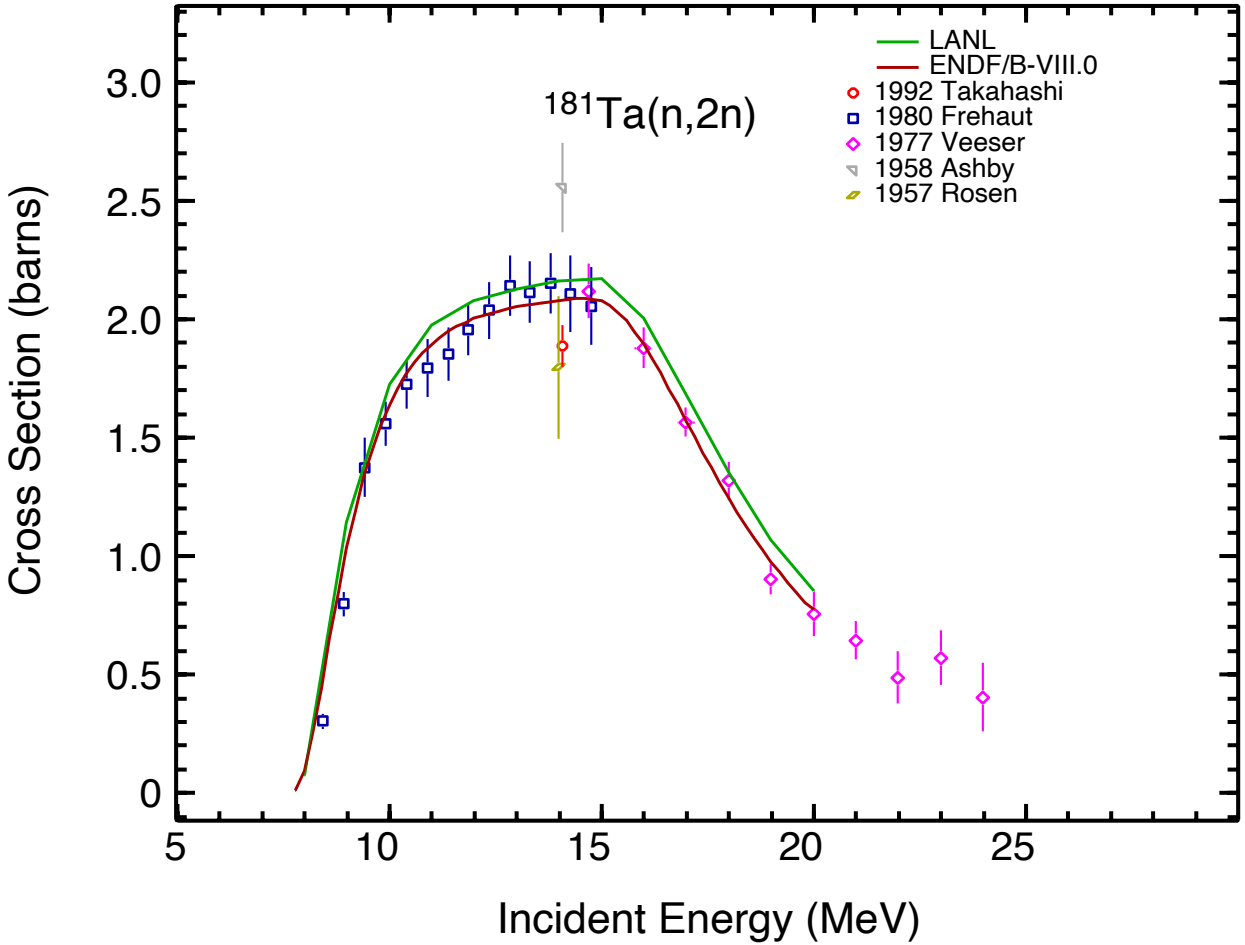
Inelastic cross sections



Ignoring data by Smith, Takahashi and Rosen as inconsistent with the overall picture.

Relatively good agreement with VIII.0.

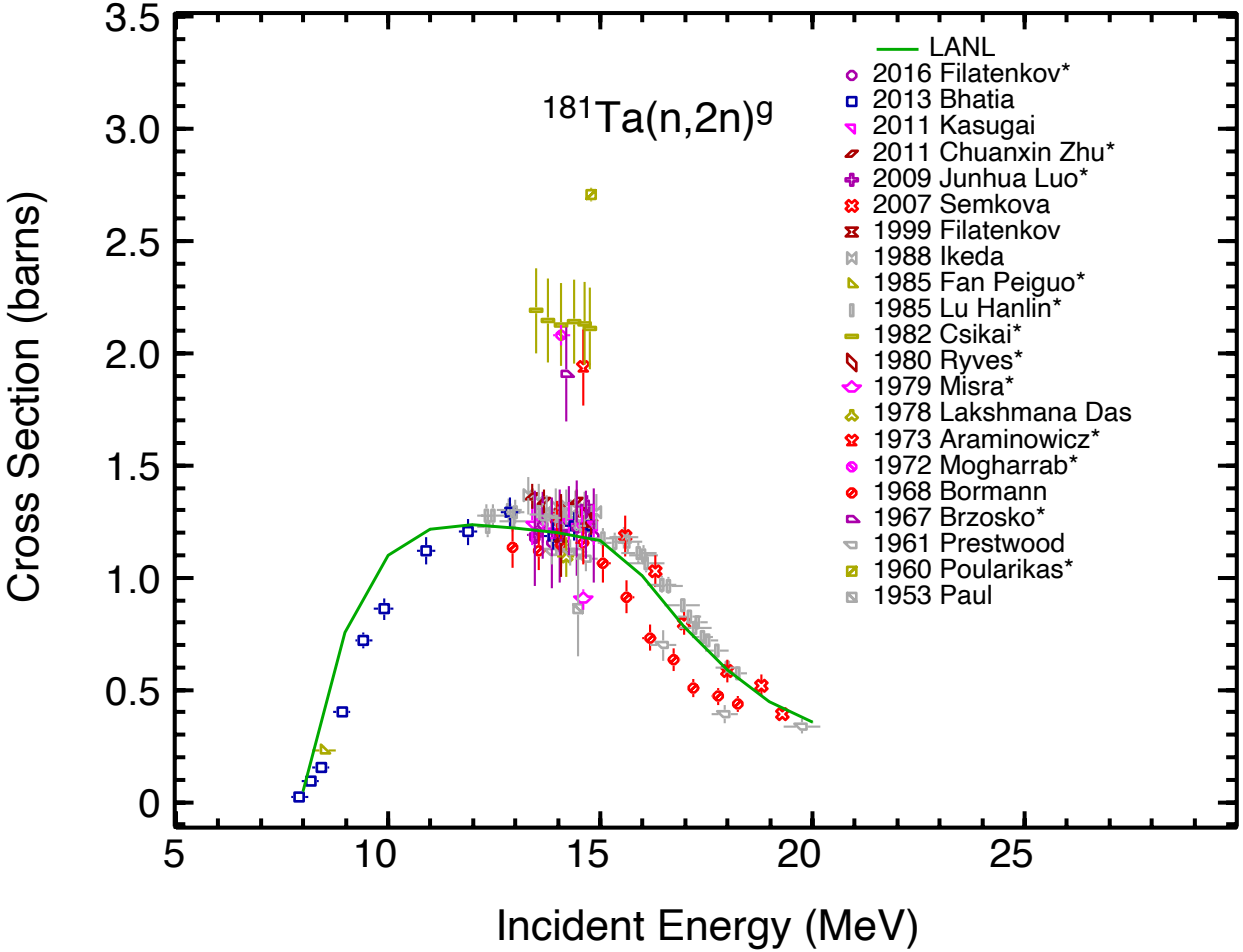
(n,2n) cross sections



VIII.0 agrees a bit better with experimental data.

The difference could be eliminated with the parameter fitting. Newly developed level densities have a substantial impact on (n,2n) cross sections.

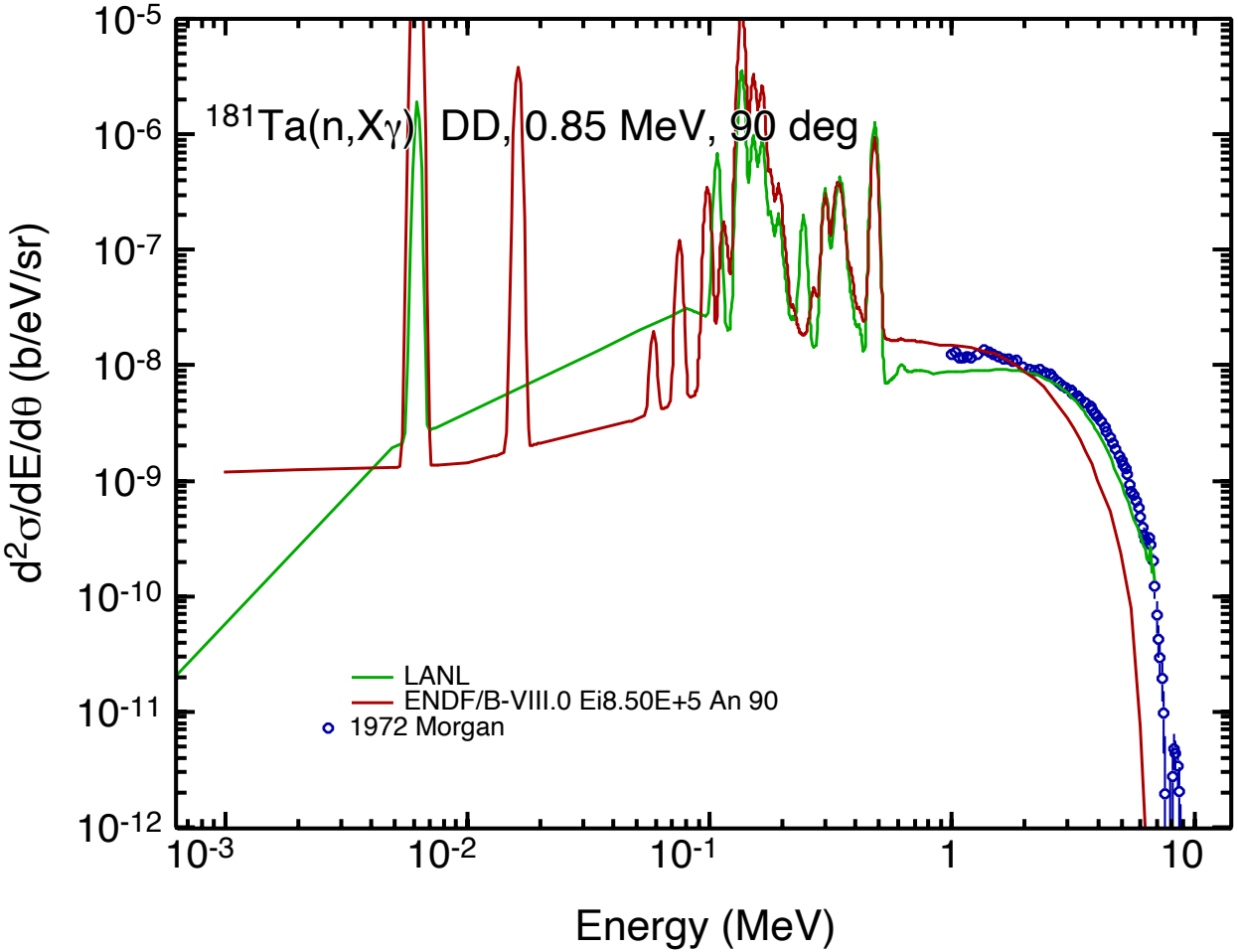
^{181}Ta(n,2n)^{179g}Ta cross sections



Agreement has been improved by completing decay schemes.

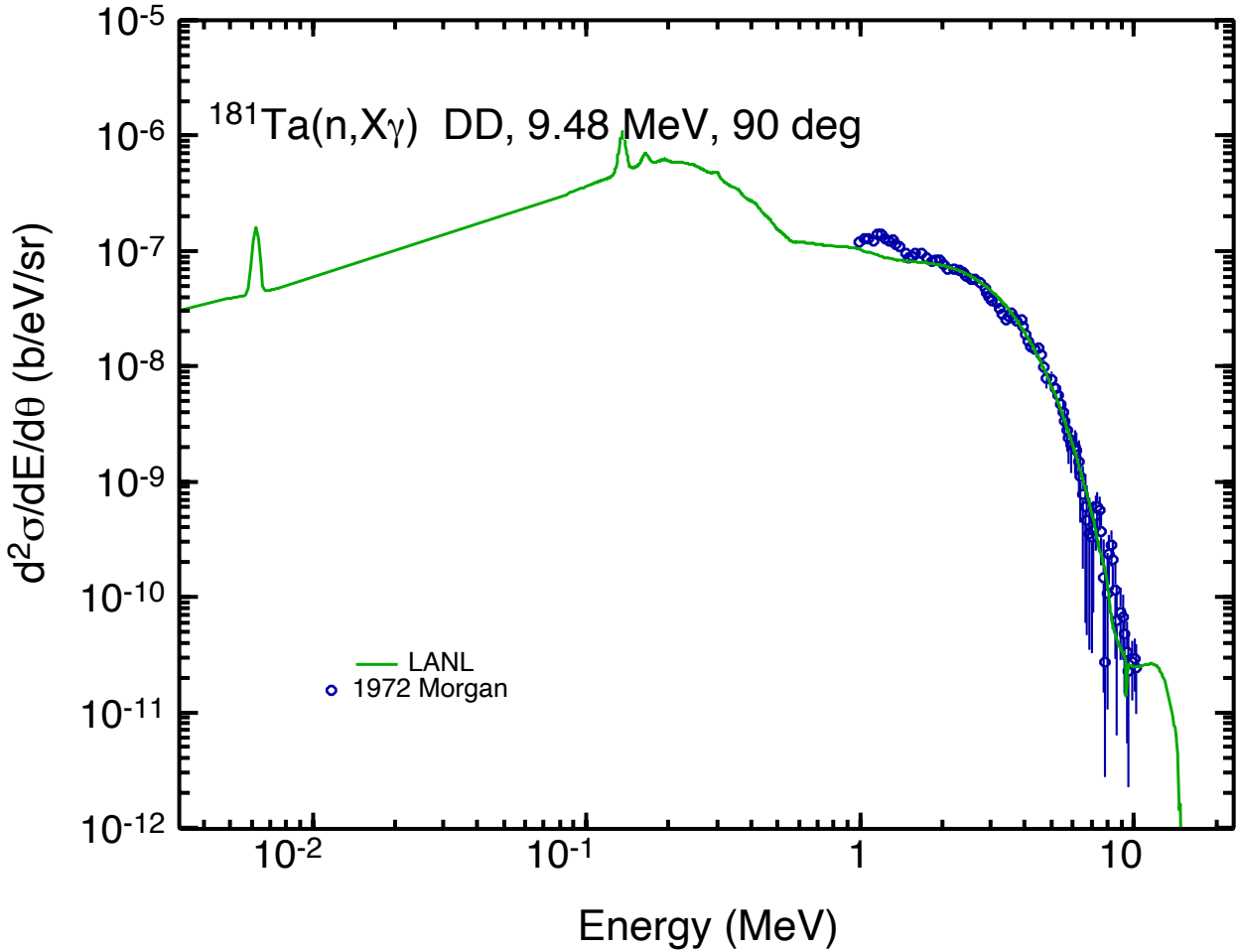
Some data need to be disregarded.

Gamma spectrum



VIII.0 has better energy resolution below 0.1 MeV but present evaluation is closer to the data above 2 MeV

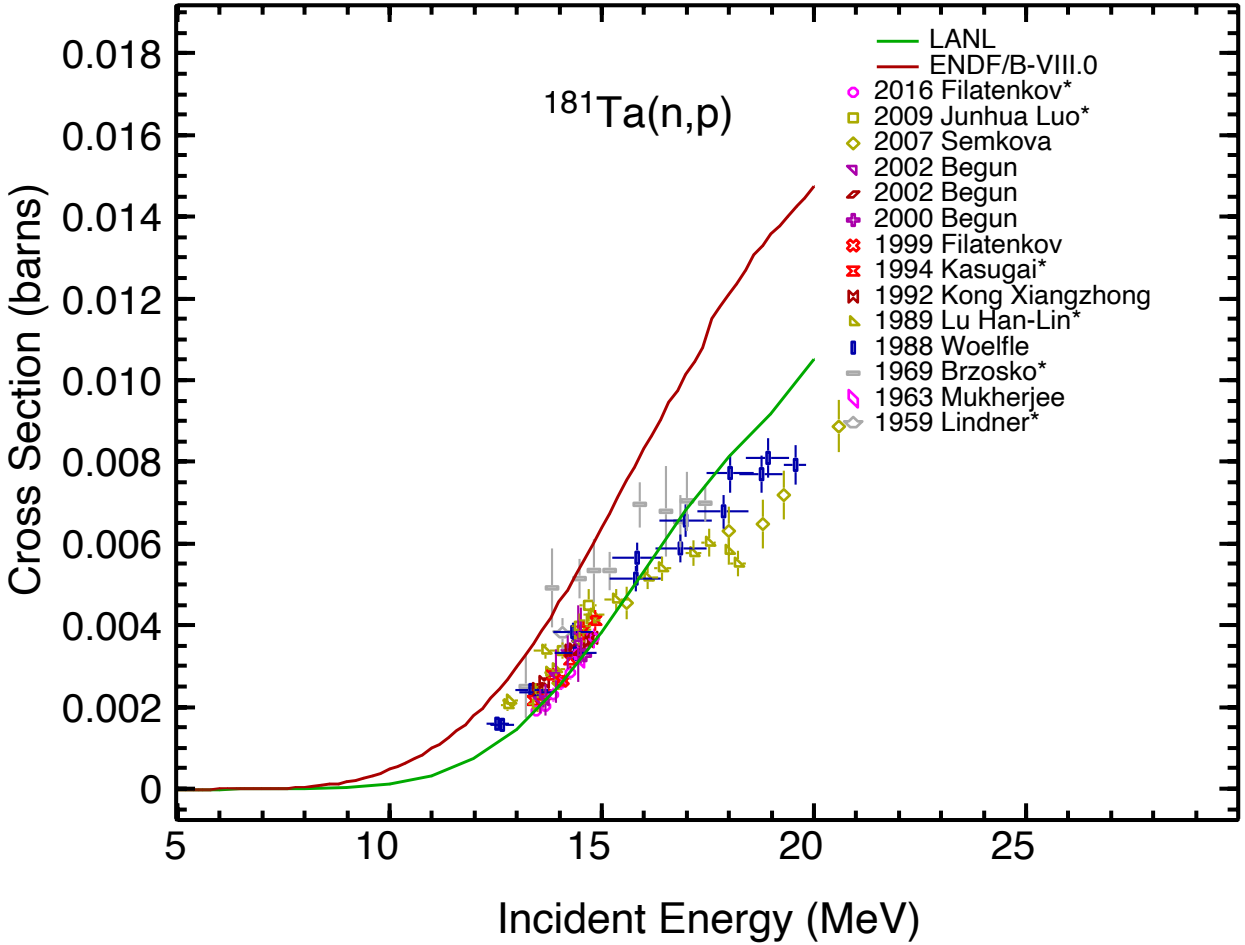
Gamma spectrum



VIII.0 gives no gamma spectra above 1 MeV of incident neutrons.

The new evaluation covers the whole energy range up to 20 MeV.

Exciton model - charged particle emission



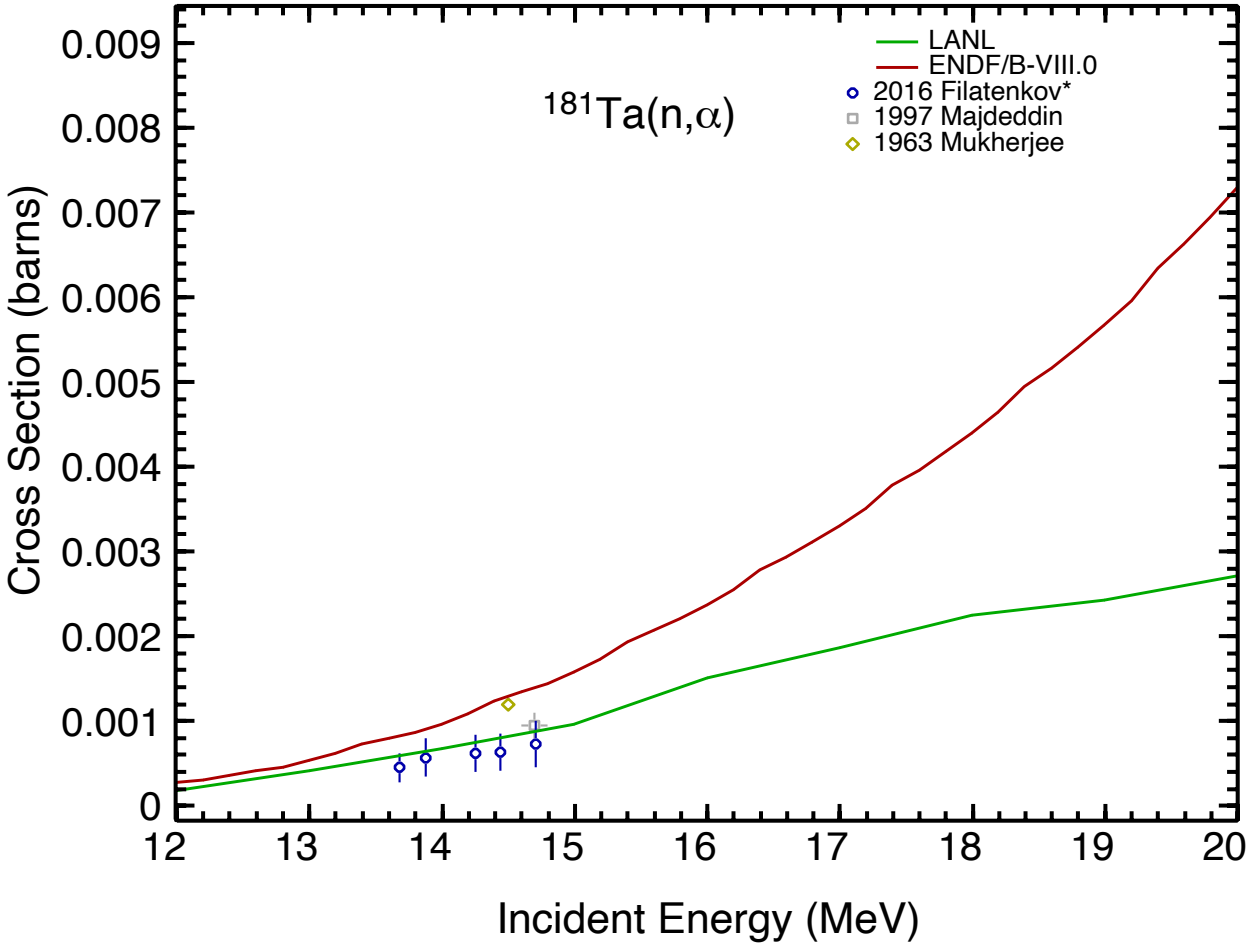
Exciton model is the only option in EMPIRE for charge-exchange reactions and cluster emission in the pre-equilibrium (PE) domain. With neutrons treated within MSD/MSC there is more freedom to adjust PE yield.

(n,p) and (n,α) reactions required reduction of the mean-free-path multiplier from the default 1.5 down to 0.8.

MFP = 0.8

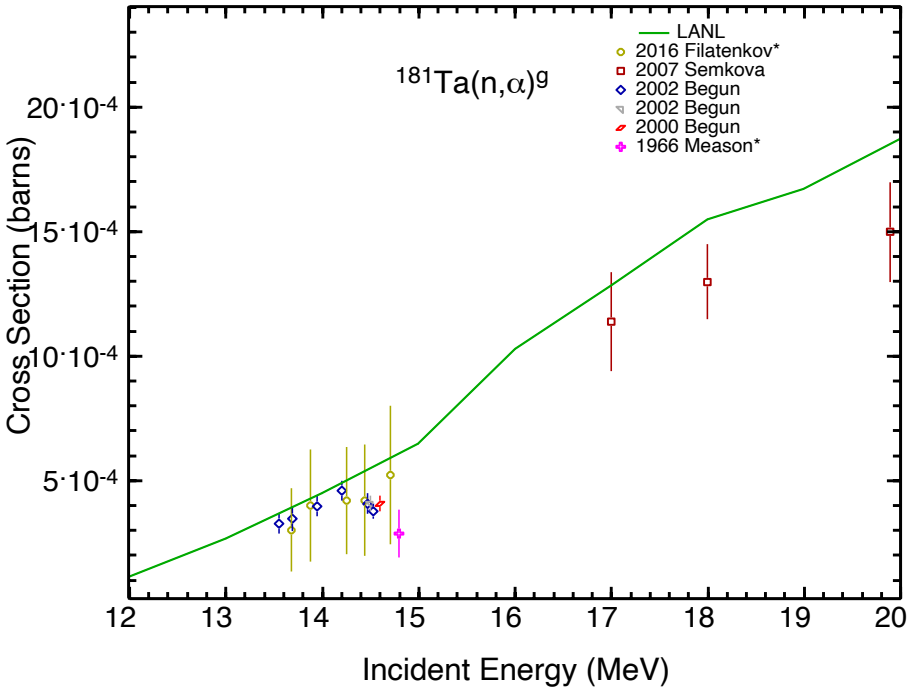
(n,p) evaluation was guided by Filatenkov's and by Semkova's data.

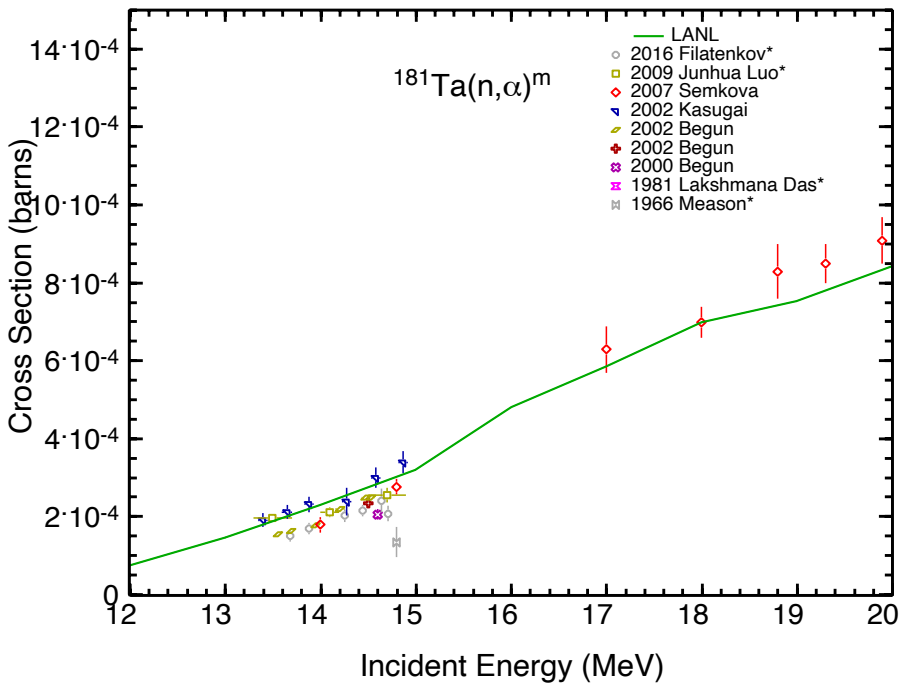
(n,α) reaction



VIII.0 was probably misguided by the old Mukherjee experiment.

Isomeric data confirm total (n,a) cross sections





Decay schemes completed to avoid default transitions to the g.s.

Modeling summary

- Coupled-Channels with adjusted dispersive Optical Model potential
- Multistep Direct (MSD) model for pre-equilibrium neutron emission.
- Heidelberg formulation of the Multistep Compound (MSC) model for pre-equilibrium neutron and γ-emission.
- Exciton model for pre-equilibrium proton emission.
- Exciton model with Iwamoto-Harada extension for pre-equilibrium cluster emission.
- Gilbert-Cameron model for level densities.
- Hauser-Feshbach with Moldauer width correction and BB angular distributions for compound nucleus decay.

Merge with the new RR/URR

- The two independent evaluations in RR/URR (D. Barry, J. Brown, M. Pigni, A. Lewis) and the present fast neutron-range evaluation show surprisingly good agreement at the 100 keV matching point
- We are working on solving a couple of mismatches inside URR
  - fast neutron capture lowered down to match the new resonance evaluation below 100 keV
  - work on increasing inelastic scattering in the URR region to match fast-neutron cross sections
- Current ENDF-6 does not allow for effective use of competitive width. Meantime…

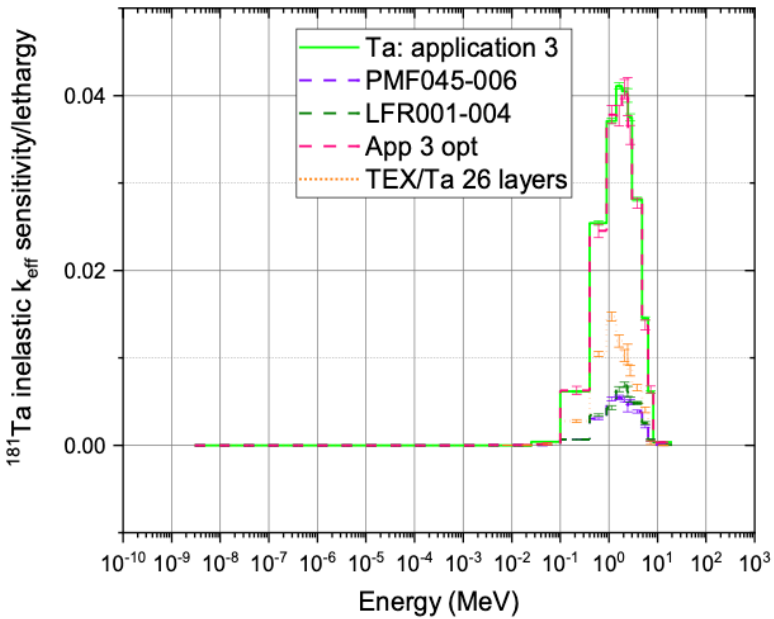
We work on both evaluations to make them fully compatible in the whole URR

Validation

- FAUST/CRATER estimates

Benchmark.	Diff. pcm	Old Bias pcm	New Bias pcm
PMF-045-001-s	-90	-388	-478
PMF-045-002-s	-94	305	211
PMF-045-003-s	-83	168	85
PMF-045-004-s	-87	167	80
PMF-045-005-s	-49	206	157
PMF-045-006-s	-87	723	636
PMF-045-007-s	-77	691	614

Much higher sensitivity with TEX



Promising trend indicated by the CRATER…

From: ARCHIMEDES LDRD

Conclusions

- Reliable fast-neutron evaluation can be entirely encapsulated in the reaction model and related input
- Ta181 is a particularly relevant case due to extraordinary coverage of various observables by differential experiments that offer much help and relatively little headache.
- Difference from the similar ENDF/B-VIII evaluation
  - more advanced modeling (CC OMP, MSD, MSC, decay schemes)
  - more careful selection of models and parameters
  - new experimental data
  - isomers
- Overall improved agreement with differential data and hints of better performance in integral testing