Update on ^{17}O evaluation and fast neutron evaluation for ^{181}Ta

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LANL/T-2

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Update on 17O system evaluation

M. Paris [M. Herman presenting]

^{17}O ~ n+^{16}O, α_0+^{13}C, n_1+$^{16}O^*$, n_2+$^{16}O^{**}$

- Continuing efforts on ^{7}Li & ^{9}Be [reported elsewhere]
- **^{17}O**
  - review of previous ENDF/B evaluations
    - ENDF/B-VI.8 (April, 2001) LANL n+^{16}O evaluation (by Chadwick, Hale & Young): evaluation done prior to Harissopulos 2005 data; ENDF/B-VI.8 is preferred by IAEA integral benchmarks
    - ENDF/B-VII.1 treated Harissopulos 2005 as "definitive"
    - JENDL-4.0 adopted ENDF/B-VII.0 (prior to Harissopulos 2005 data), which is equivalent to VI.8
  - added inelastic $^{16}O^*(0^+$; 6.05 MeV) and $^{16}O^{**}(3^-$; 6.13 MeV) inelastic channels to EDAf90 R-matrix configurations
  - augmented data deck:
    - more complete set of ^{16}O(n,el) angular distributions and polarization data
    - $^{16}O(n,n_2)$ inelastic angular distributions
    - including $^{13}C(\alpha,n_0)$ and $^{13}C(\alpha,n_1)$ angular distributions 2021 data from Notre Dame
    - comparison with LENZ(2017) data from LANL (by Kuvin & Lee) with ENDF/B-VIII.0 parameters: good agreement below 6.25 MeV
    - new, preliminary evaluation compared with 2021 OU (Brandenburg & Meisel) and 1973 Bair & Haas data: significant improvement above E_α > 5.0 MeV (previous upper limit)
  - performed consistency study of low energy (α,n_0) data
  - presently incorporating any missing data and investigating normalizations of Cierjacks' '68 and '83 datasets
  - writing ^{17}O system paper for Phys. Rev. C publication

^{181}Ta - fast neutron evaluation

M.Herman & T. Kawano

… 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. ”

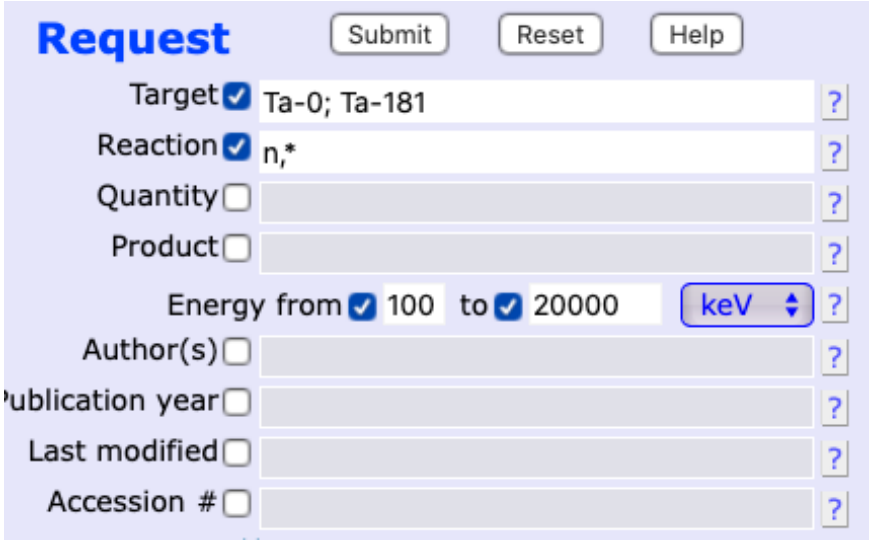
- Tantalum is used for casting of plutonium peats at LANL PF-4. k_eff for this casting is 2-3 times more sensitive to inelastic, elastic and capture on Ta than to the respective reactions on Pu239 (LA-UR-19-31175)

- 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

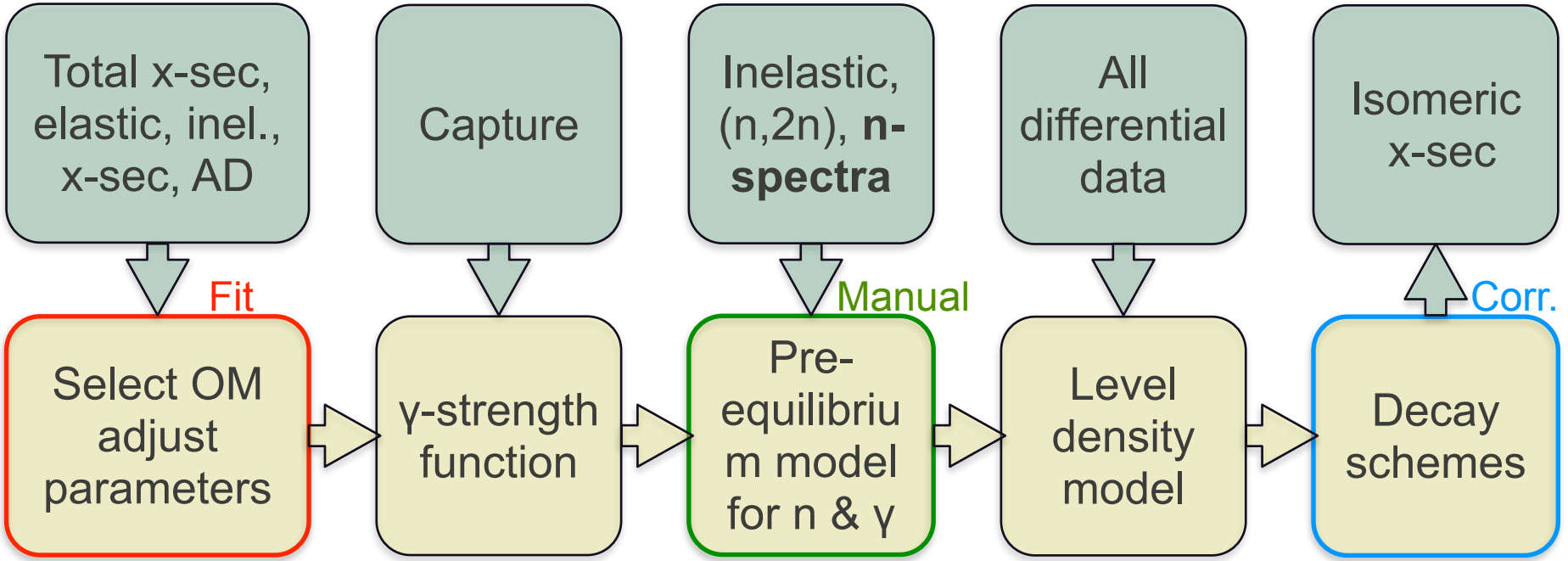


- Number of EXFOR datasets : 343
- Number of EXFOR quantities: 137
- EMPIRE makes 468 plots (!)

Extraordinarily well distributed measurements!

Concept of this evaluation

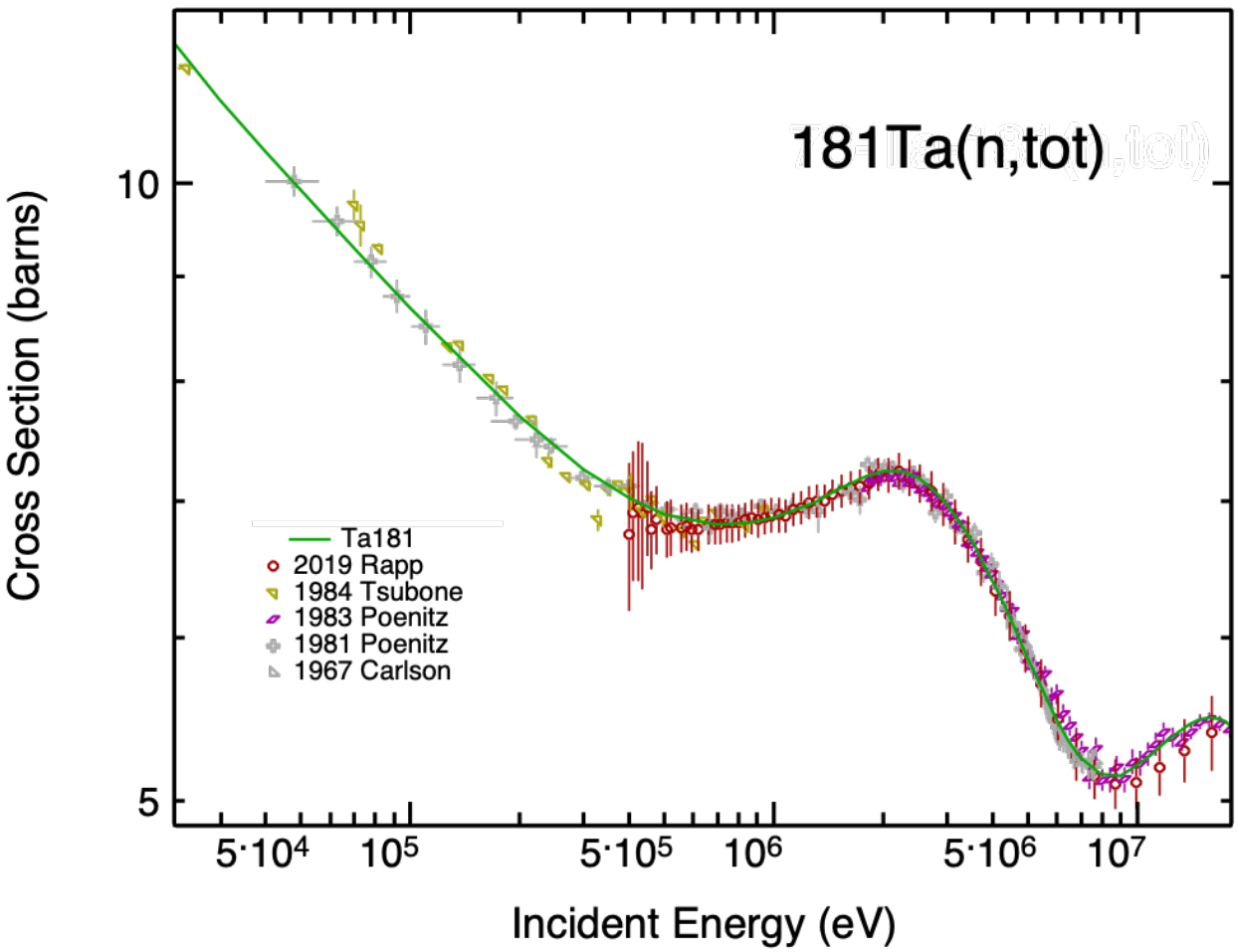
- Combine modeling of EMPIRE-3.2 with experimental data to produce a consistent picture of nuclear reactions considering “all” available differential and integral data.



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

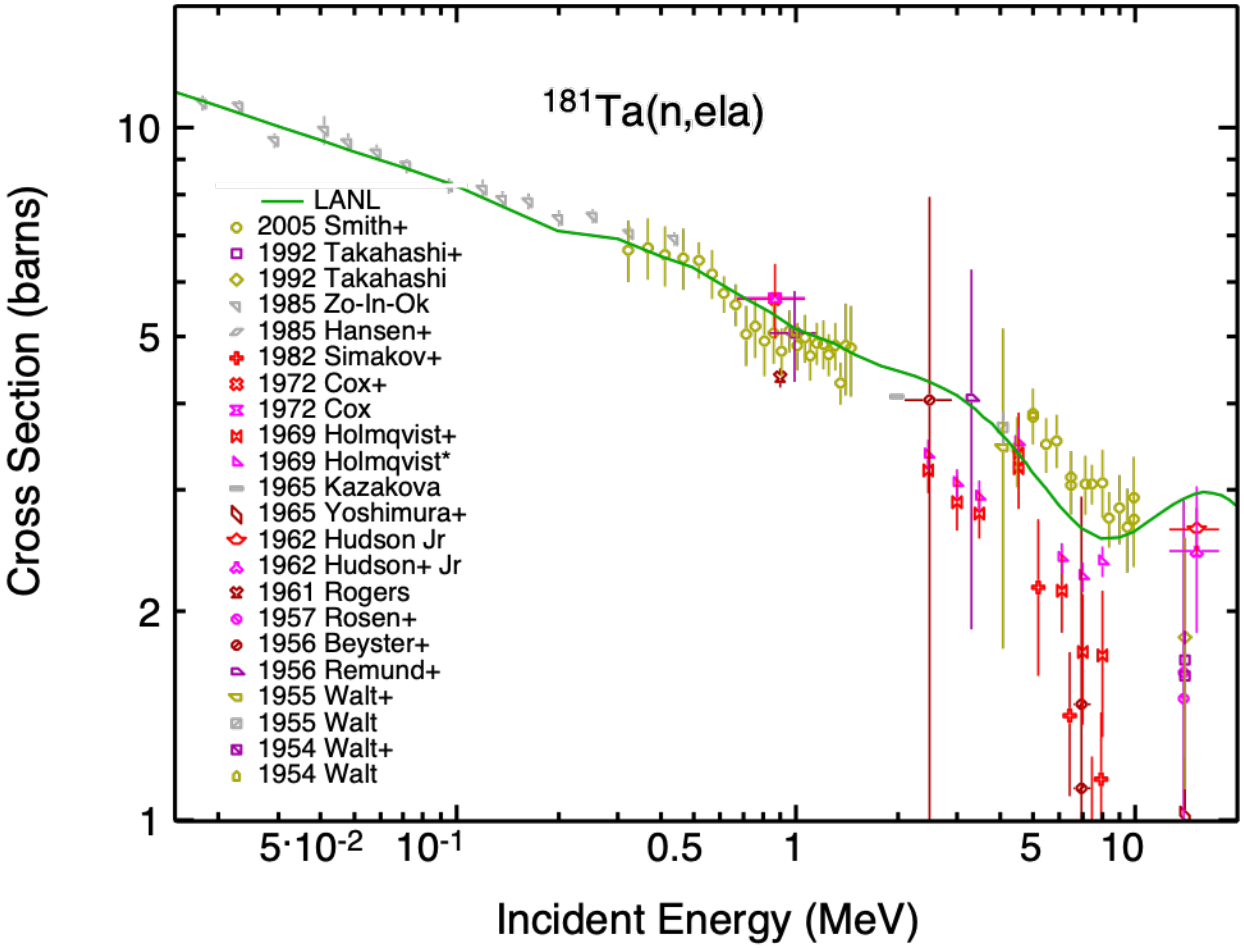
r_v	0.98542
V_v	1.0184
a_v	1.0870
β_2	1.0444
β_4	0.93707
β_6	0.49351



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

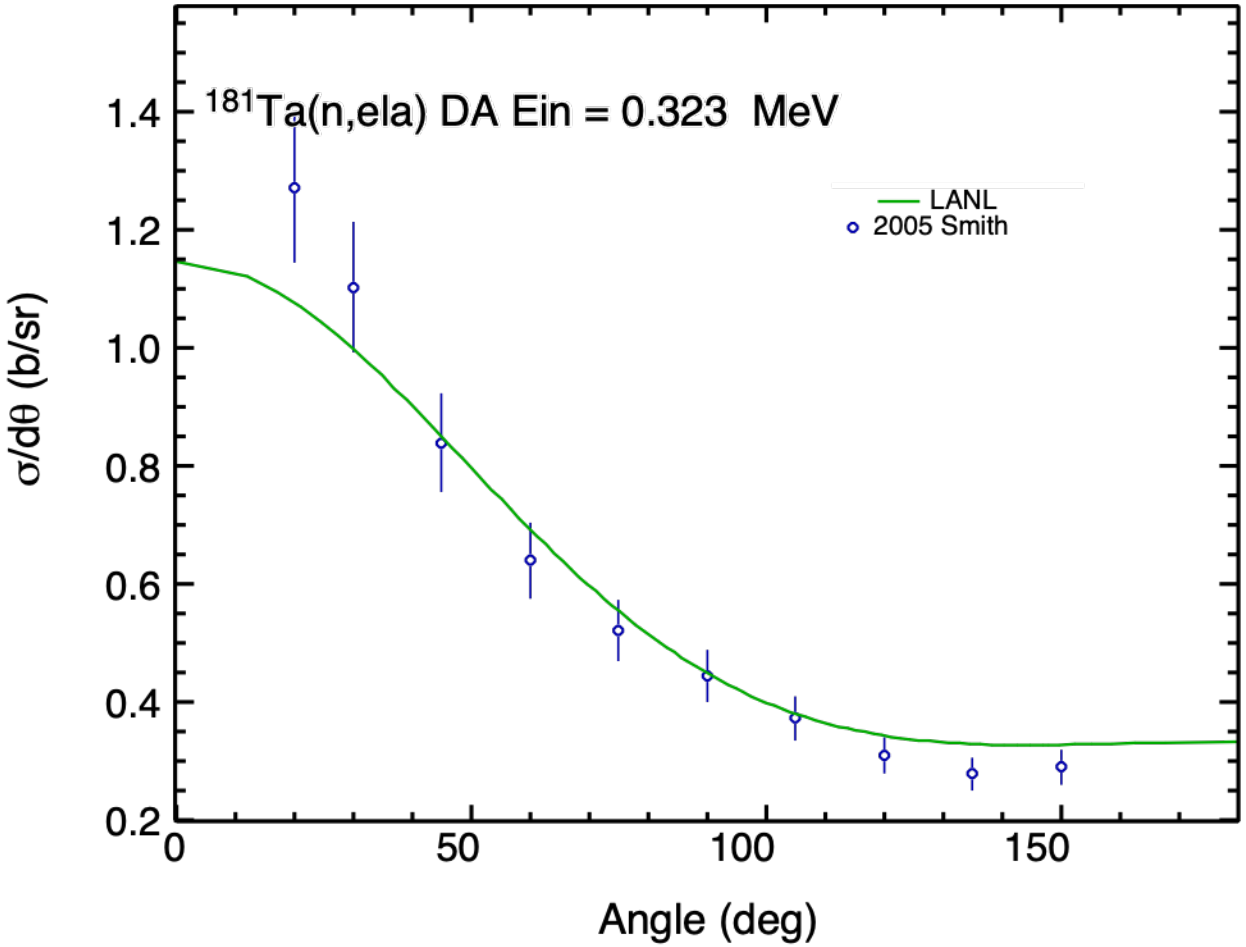
LANL evaluation perfectly fits Rapp, Poenitz 81 & 83, Tsubone, Smith, and Carlson.

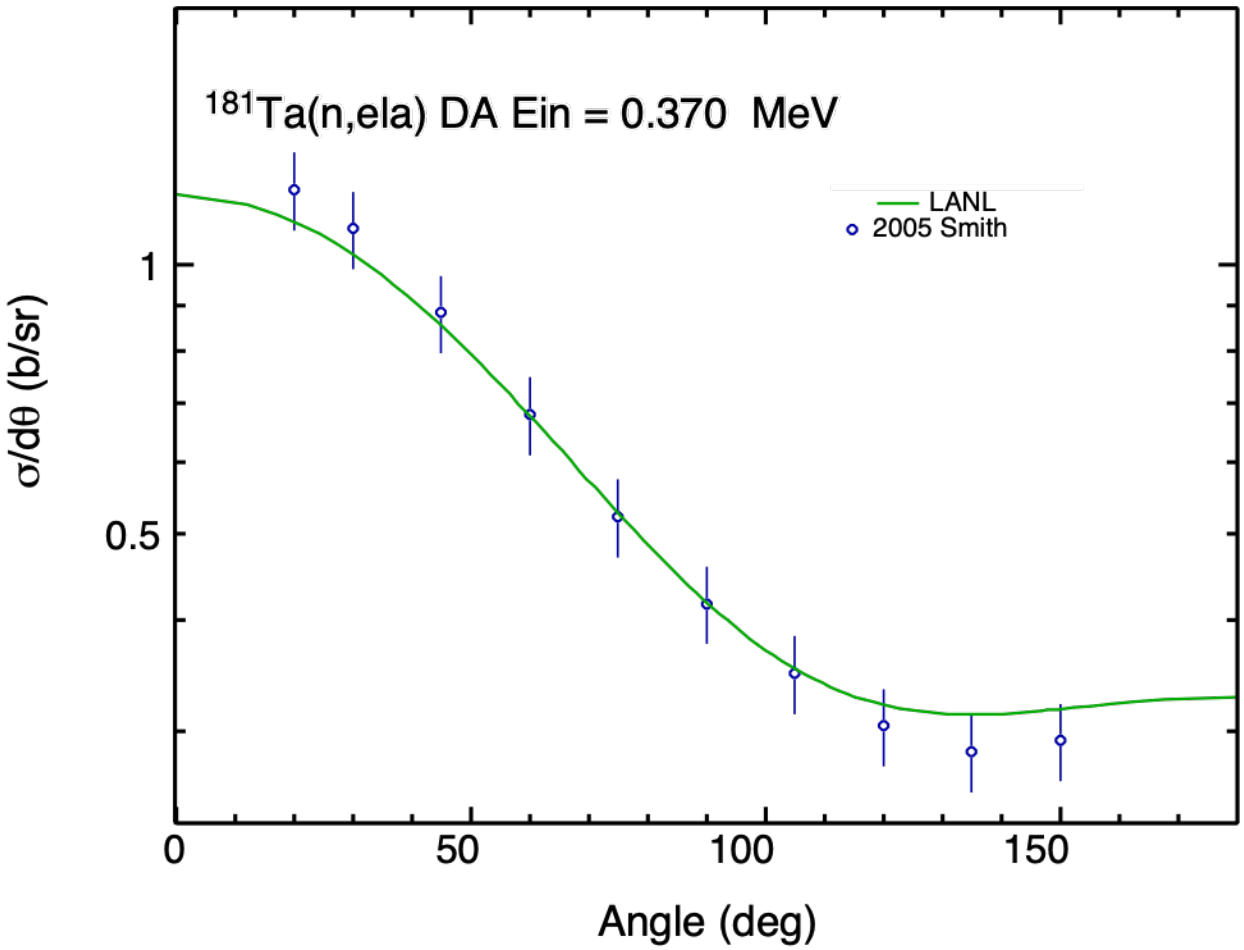
CC model - elastic x-sec & angular distributions



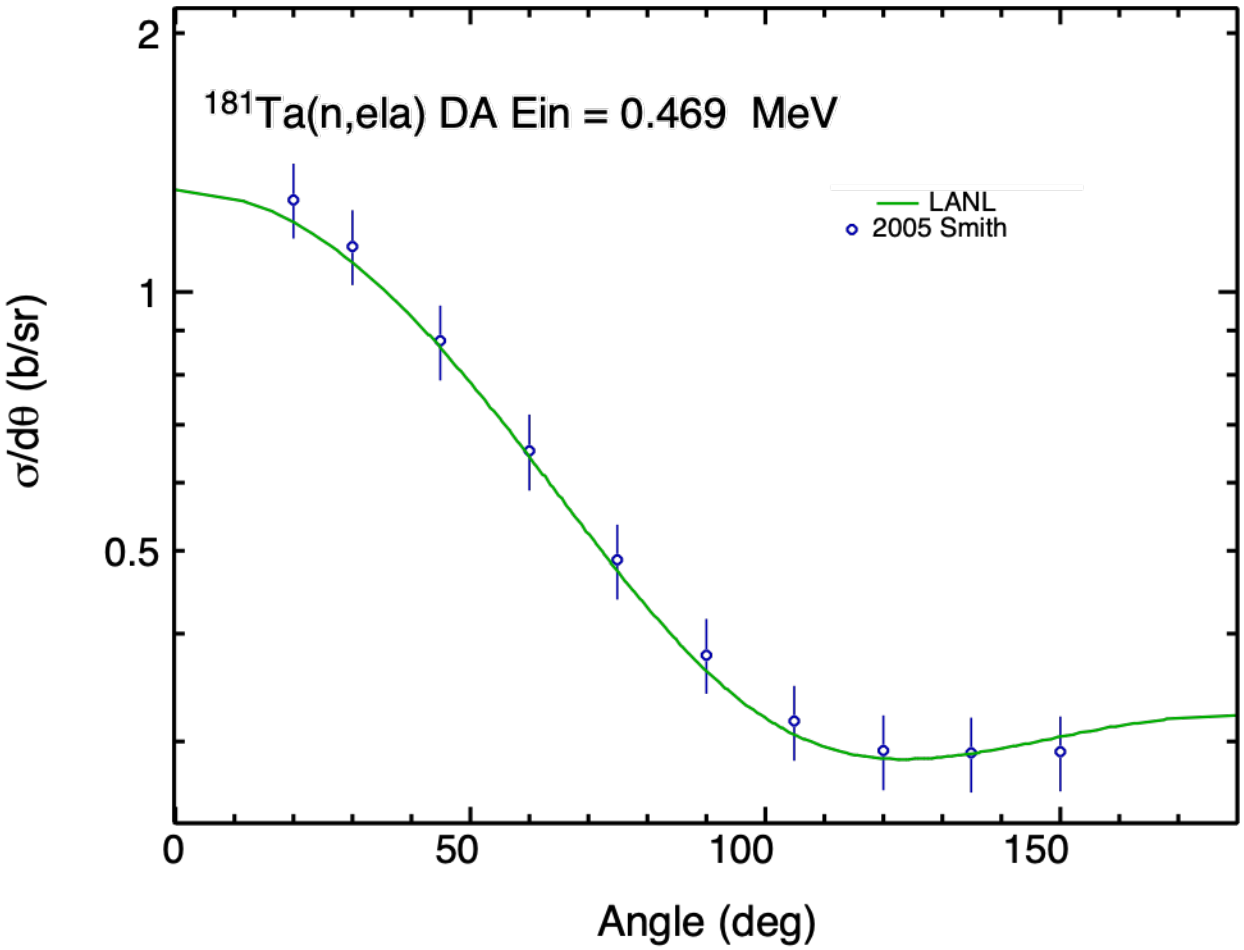
Elastic cross sections after adding the 1-st and the 2-nd inelastic.

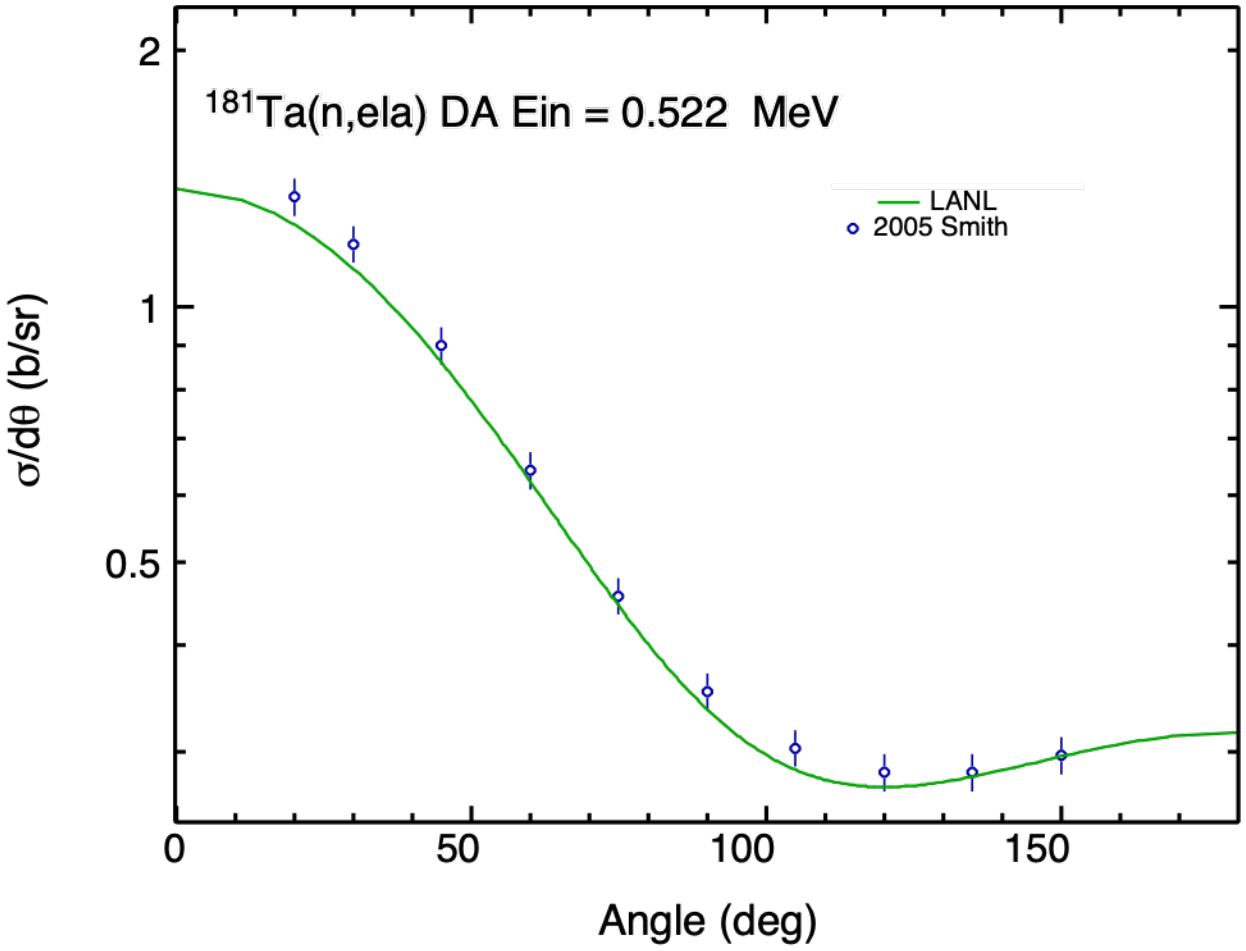
Elastic angular distributions A. Smith: 0.3 - 1.5 MeV

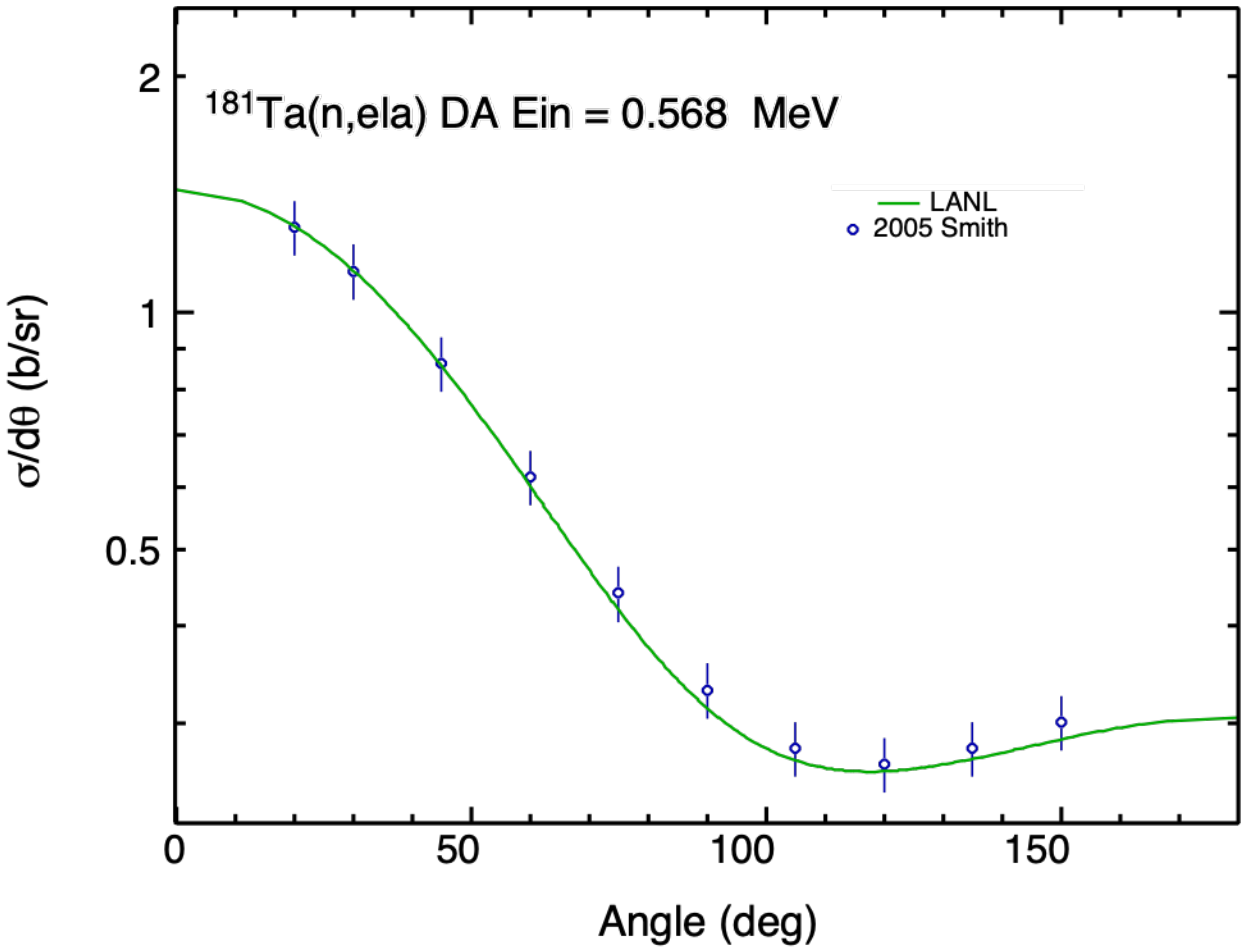


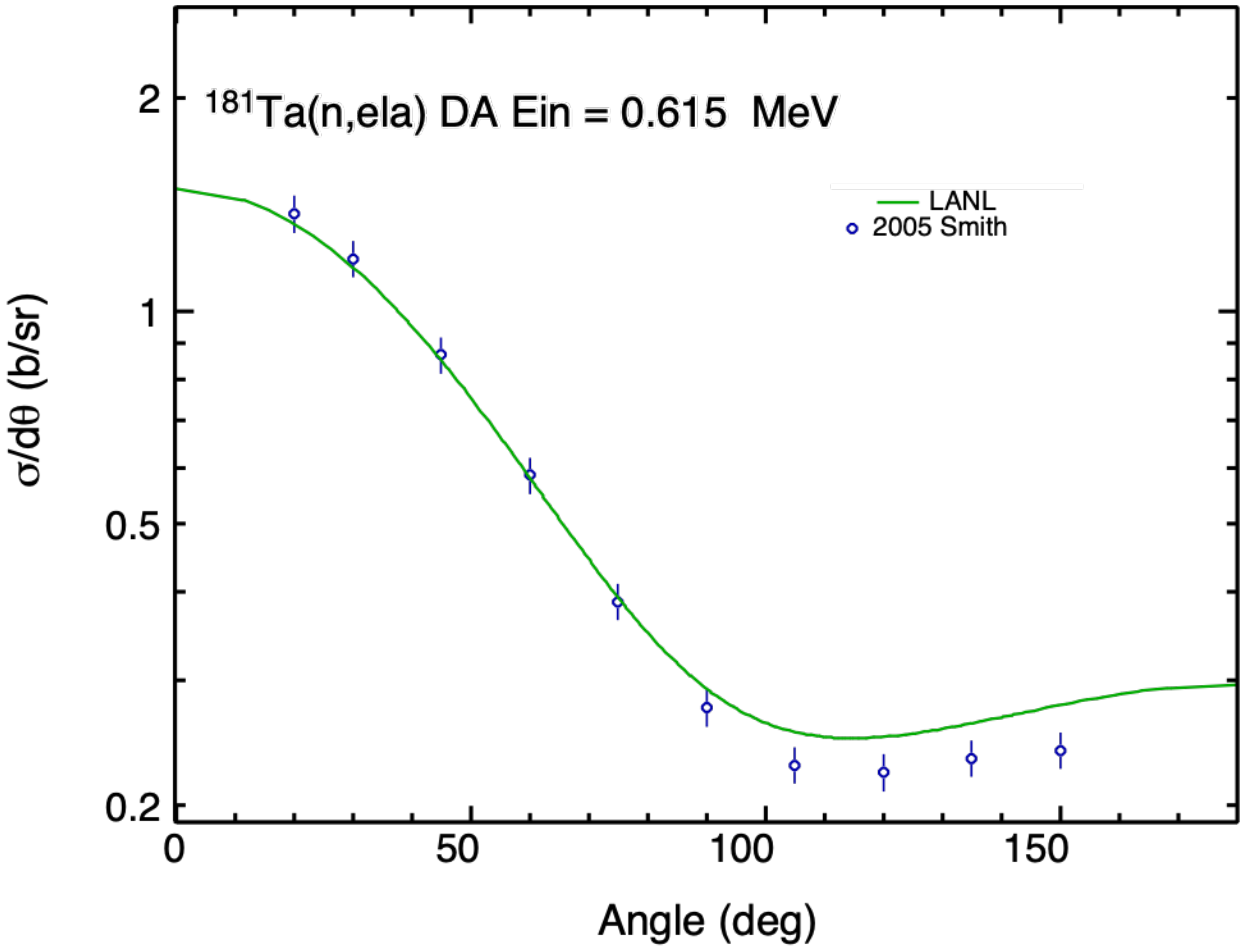


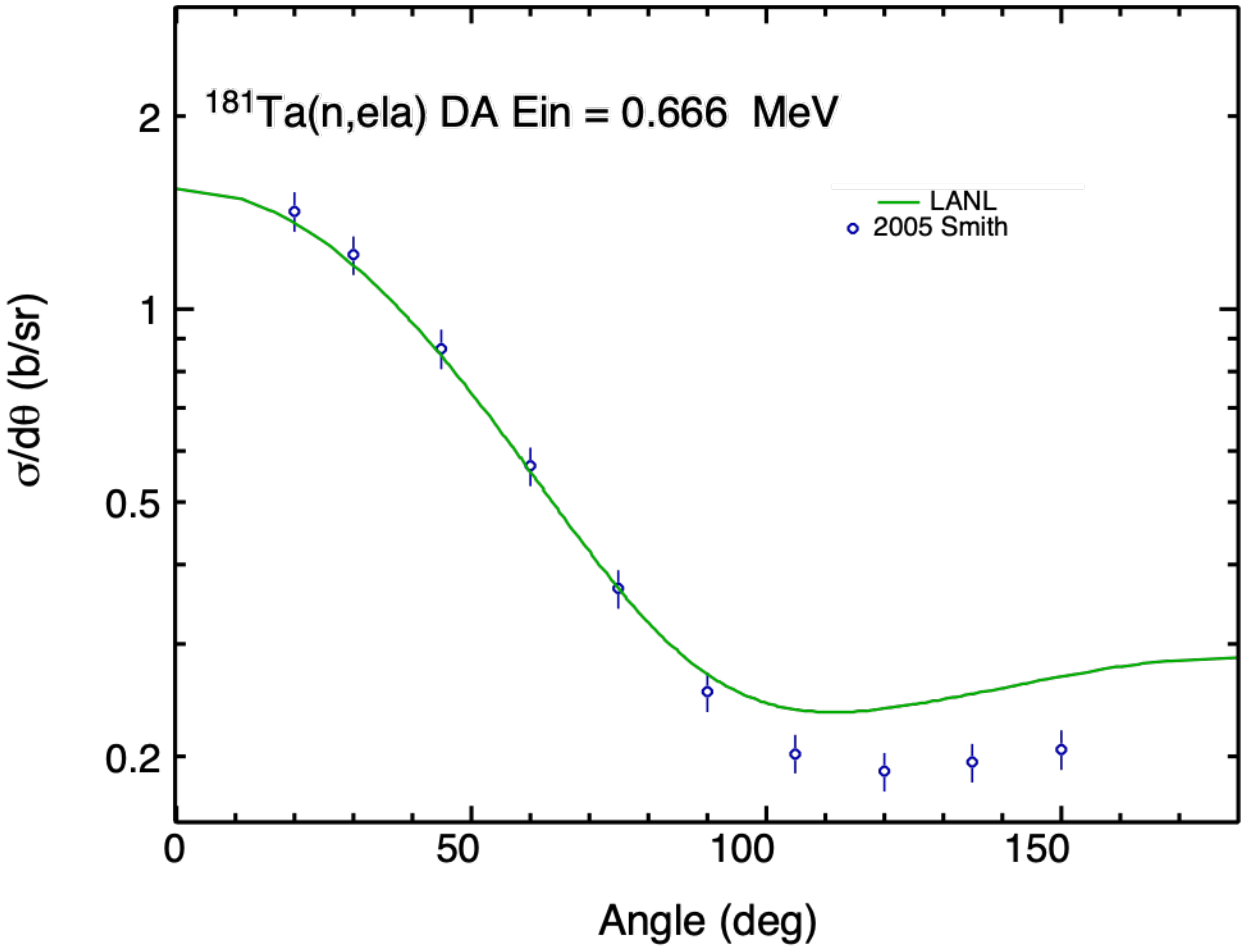
The 1-st and the 2-nd inelastic angular distributions added.

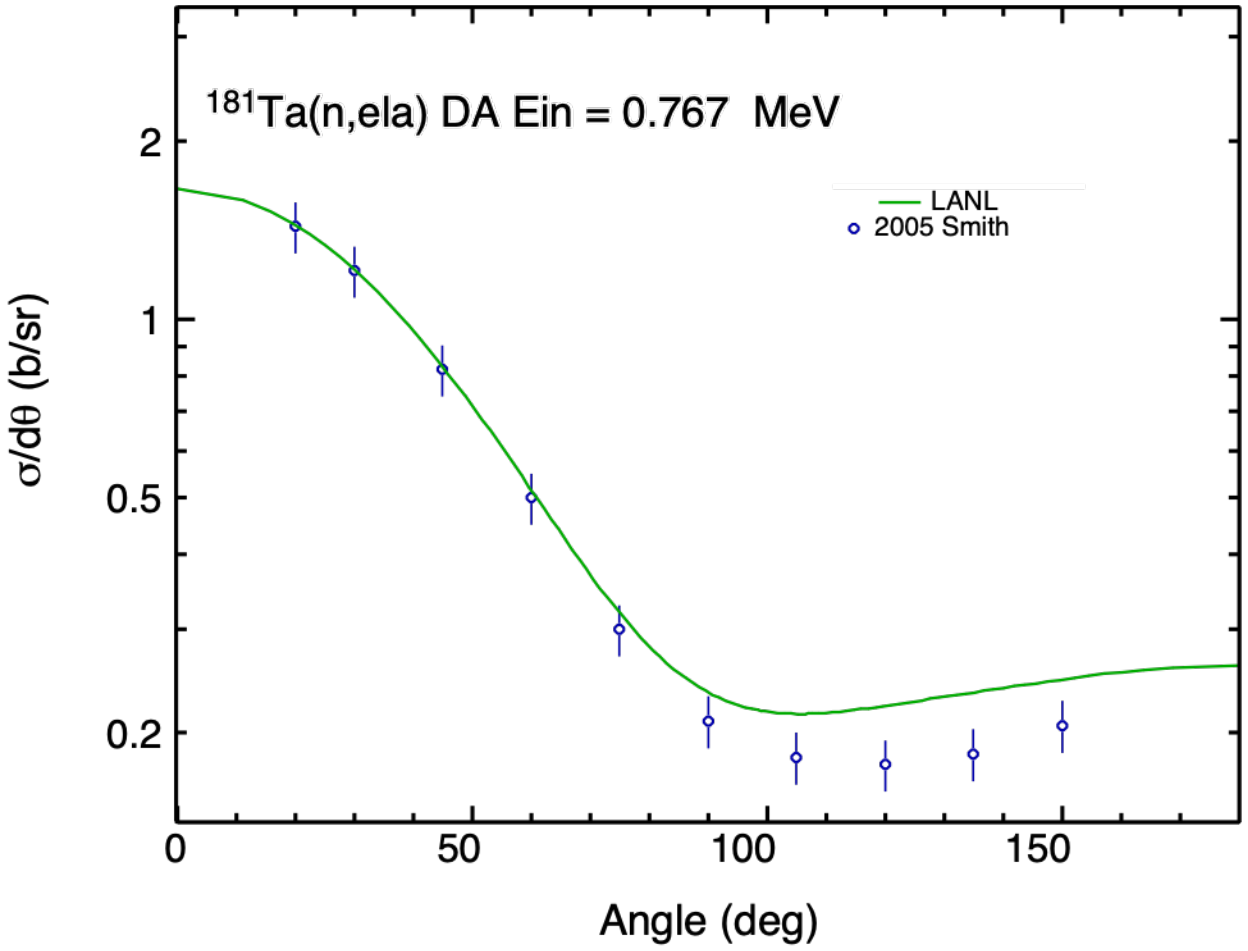

^{181}Ta(n,ela) DA Ein = 0.469 MeV
LANL
2005 Smith
σ/dθ (b/sr)
Angle (deg)
0
0.5
1
2
0
50
100
150

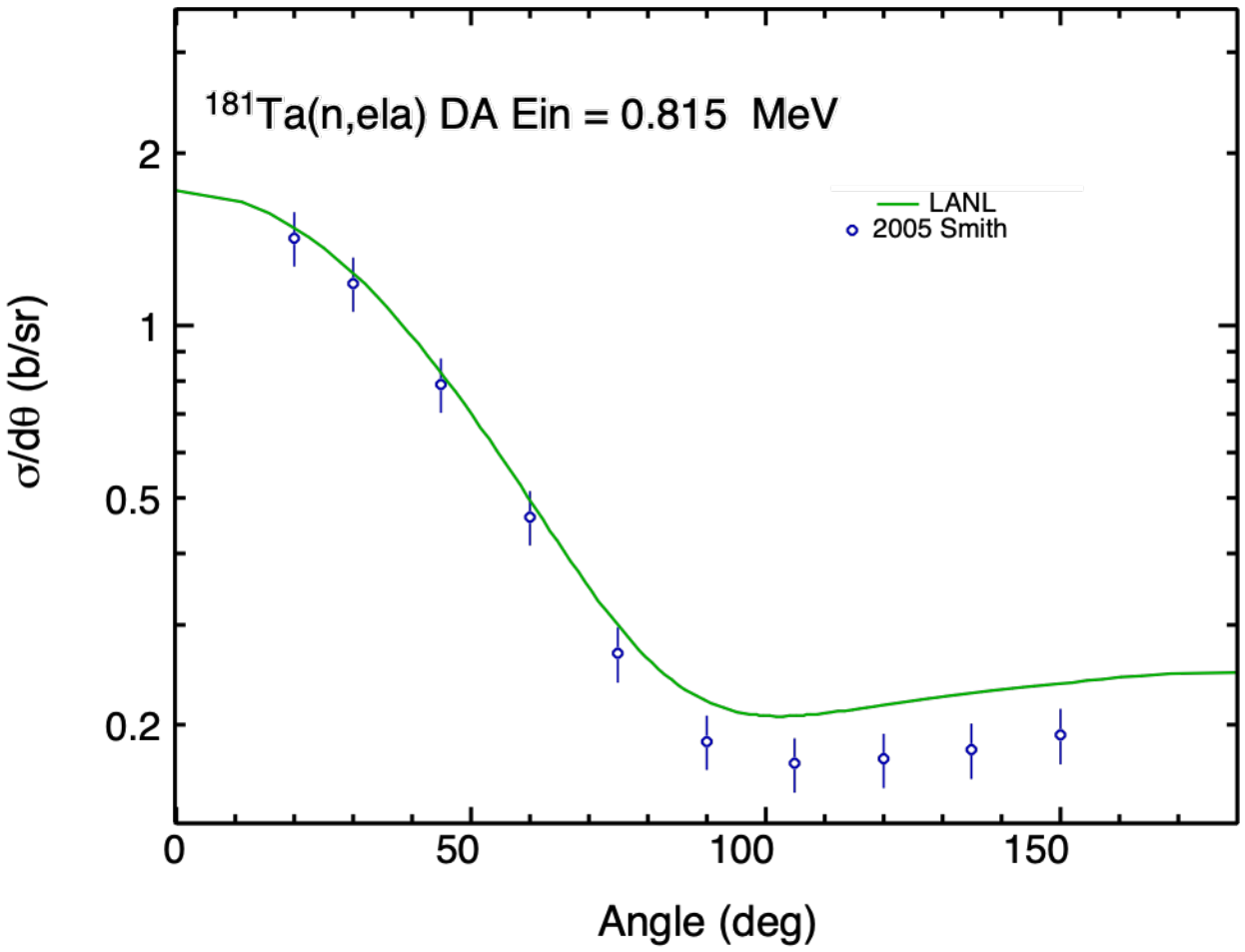

^{181}Ta(n,ela) DA Ein = 0.522 MeV
LANL
2005 Smith
σ/dθ (b/sr)
Angle (deg)
0
50
100
150
0.5
1
2

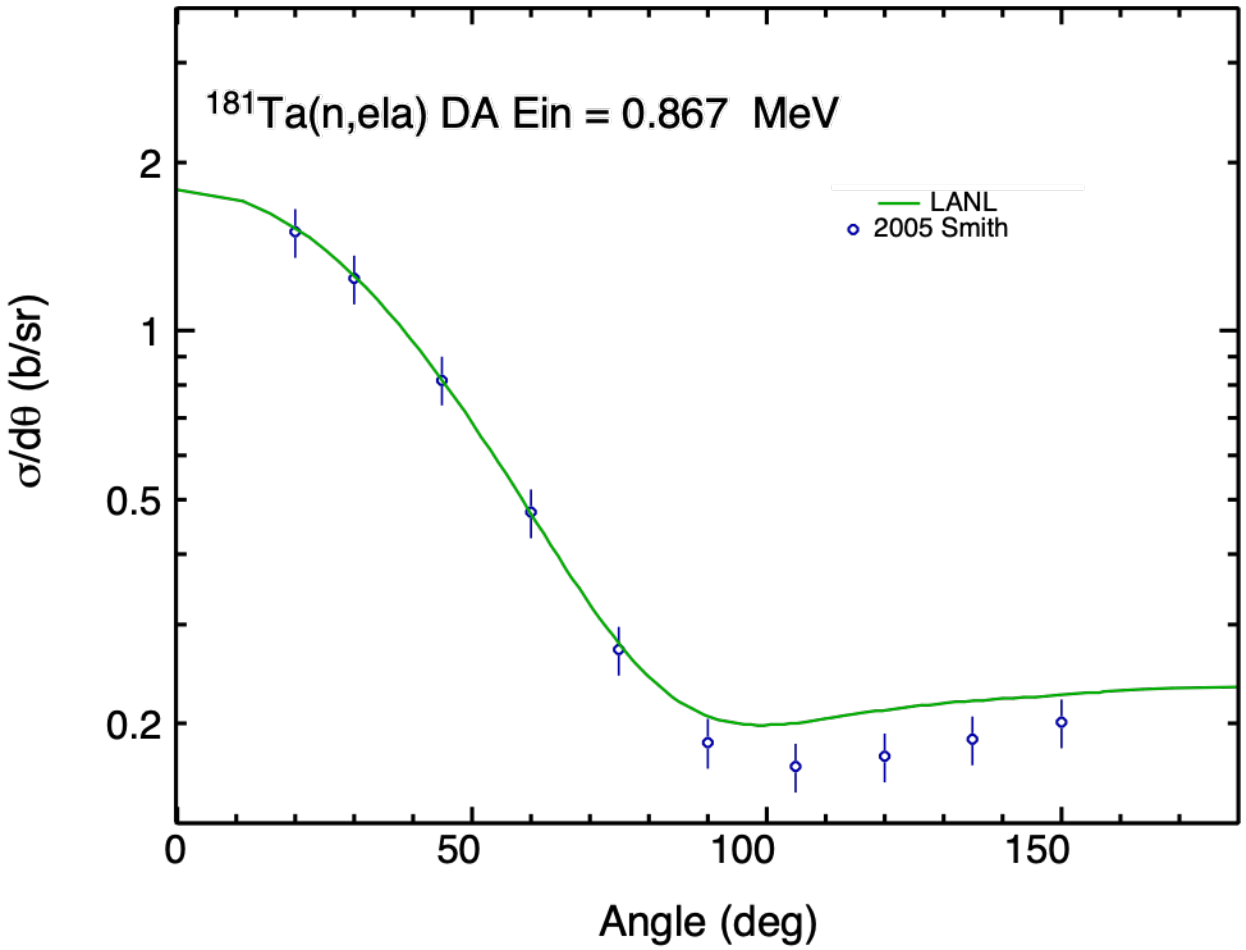

^{181}Ta(n,ela) DA Ein = 0.568 MeV
LANL
2005 Smith
σ/dθ (b/sr)
Angle (deg)
0
50
100
150
0.5
1
2

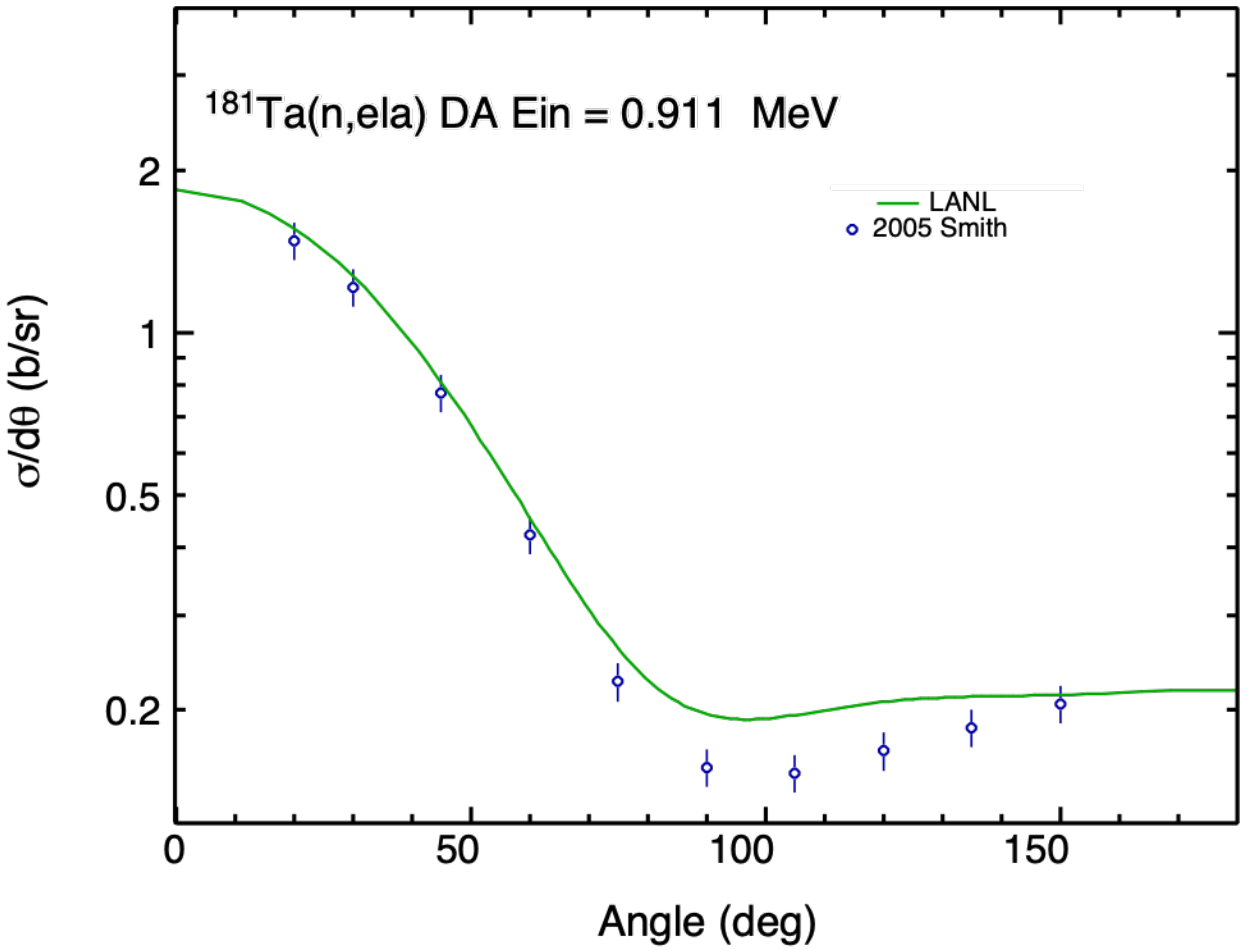

^{181}Ta(n,ela) DA Ein = 0.615 MeV
LANL
2005 Smith
σ/dθ (b/sr)
Angle (deg)
2
1
0.5
0.2
0
50
100
150

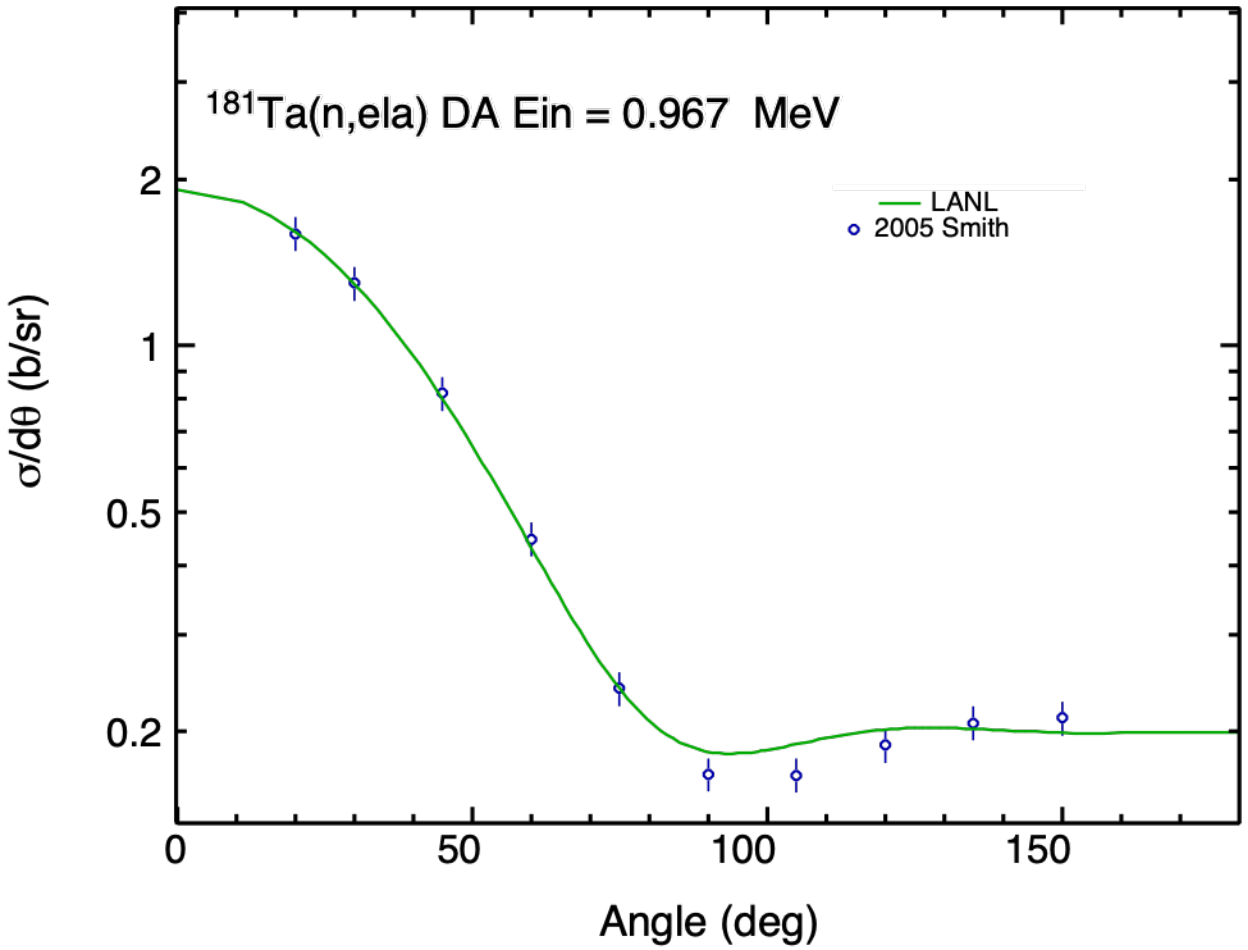

^{181}Ta(n,ela) DA Ein = 0.666 MeV
LANL
2005 Smith
σ/dθ (b/sr)
2
1
0.5
0.2
0
50
100
150
Angle (deg)

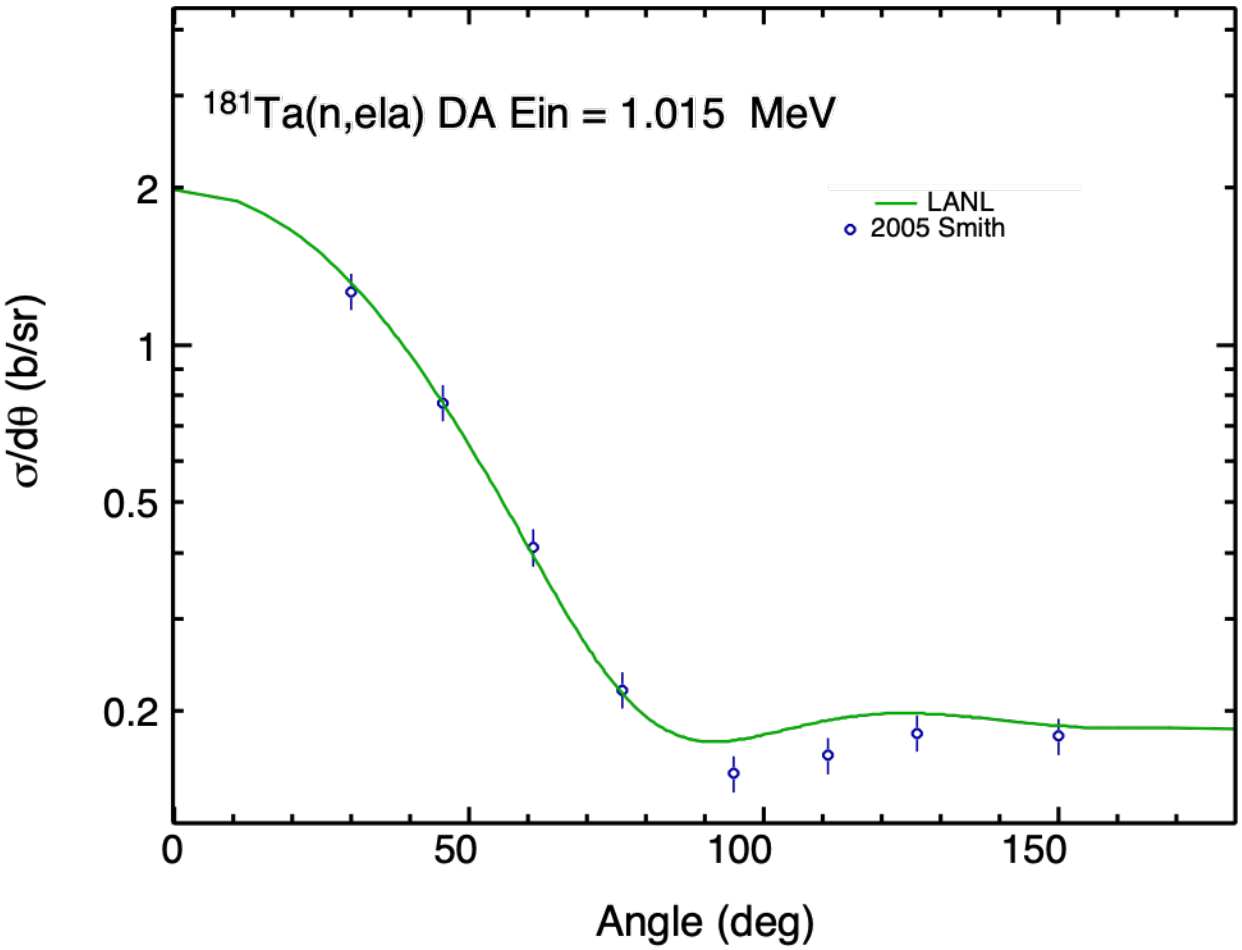

^{181}Ta(n,ela) DA Ein = 0.767 MeV
LANL
2005 Smith
σ/dθ (b/sr)
2
1
0.5
0.2
0
50
100
150
Angle (deg)

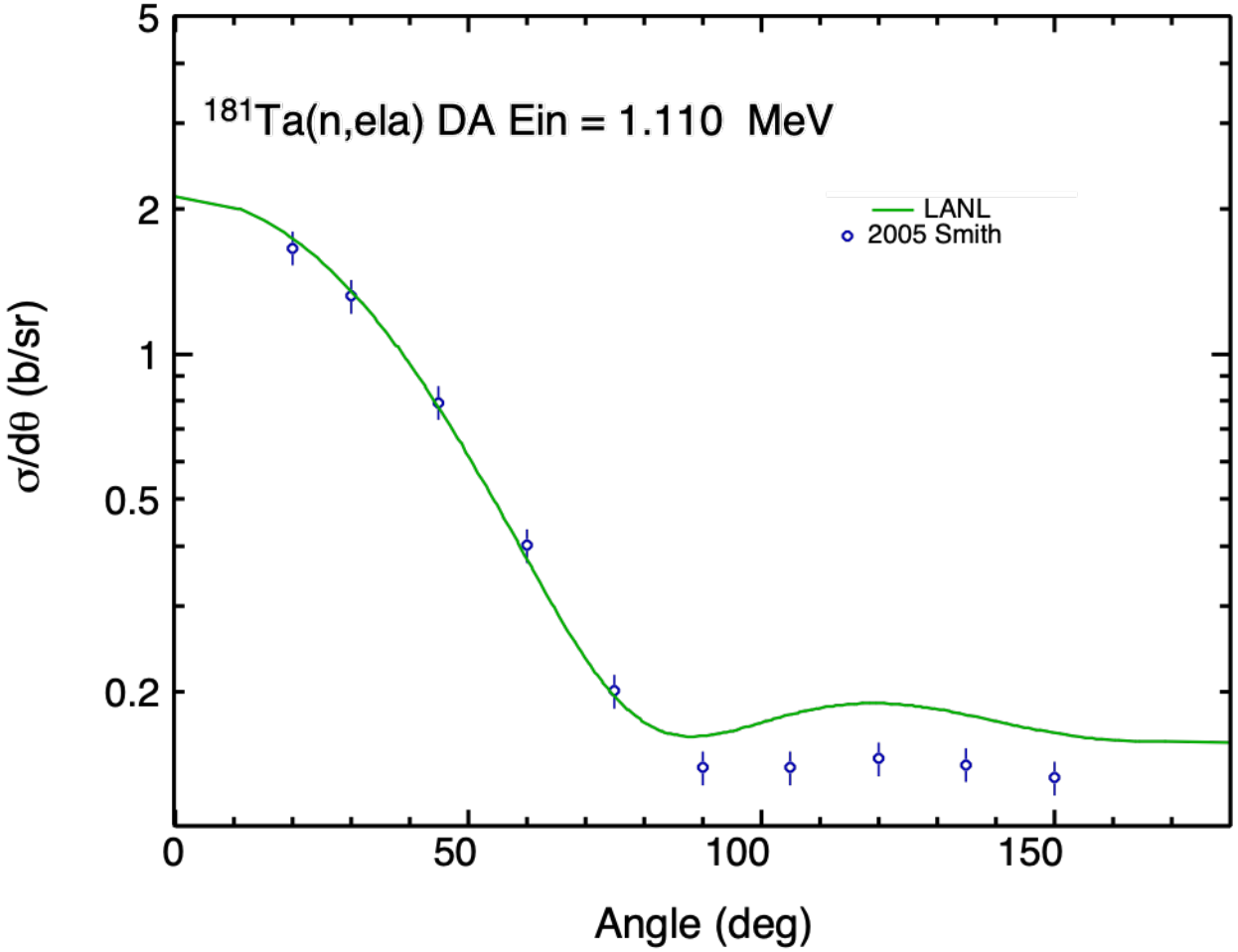

^{181}Ta(n,ela) DA Ein = 0.815 MeV
LANL
2005 Smith
σ/dθ (b/sr)
2
1
0.5
0.2
0
50
100
150
Angle (deg)

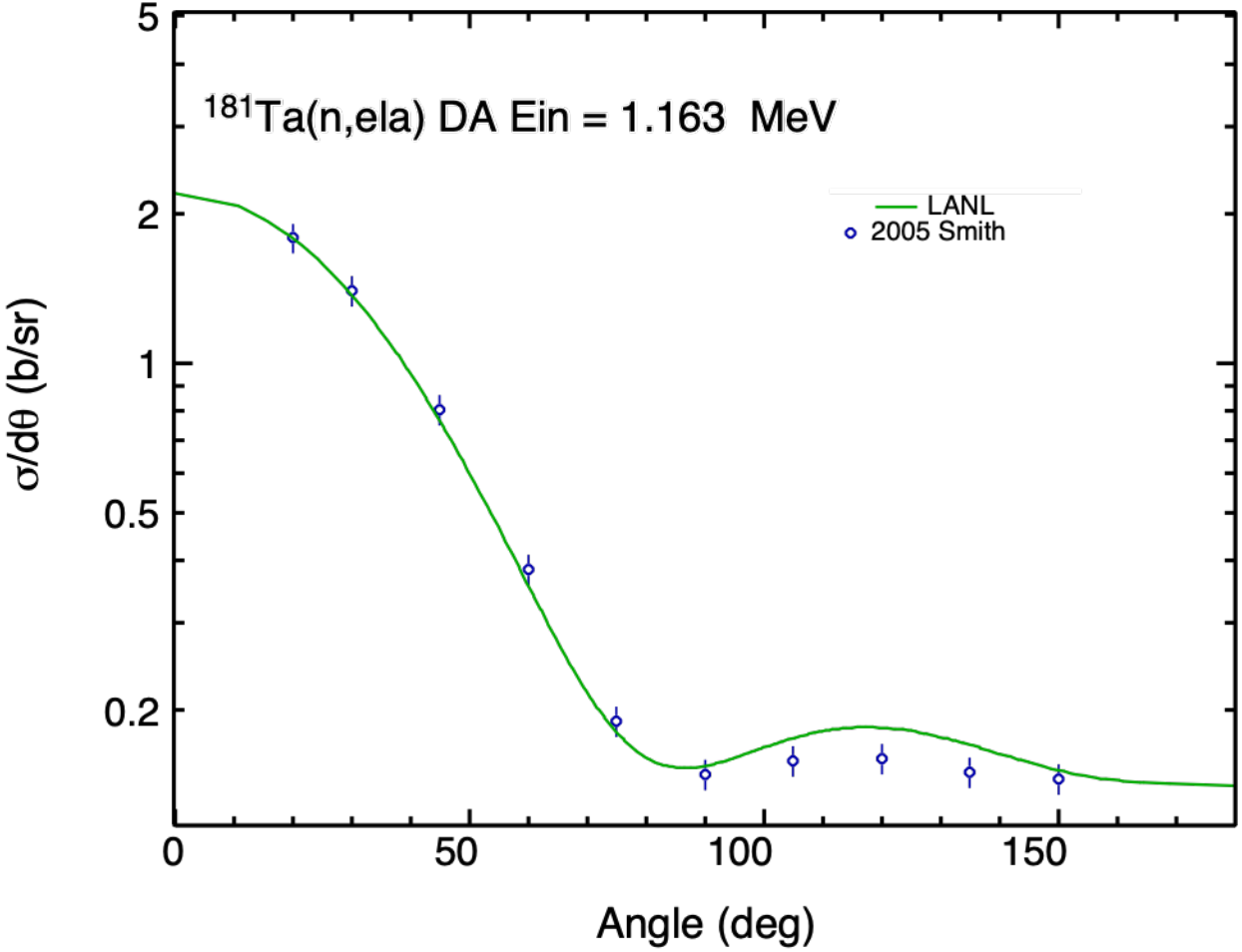

^{181}Ta(n,ela) DA Ein = 0.867 MeV
LANL
2005 Smith
σ/dθ (b/sr)
2
1
0.5
0.2
0
50
100
150
Angle (deg)

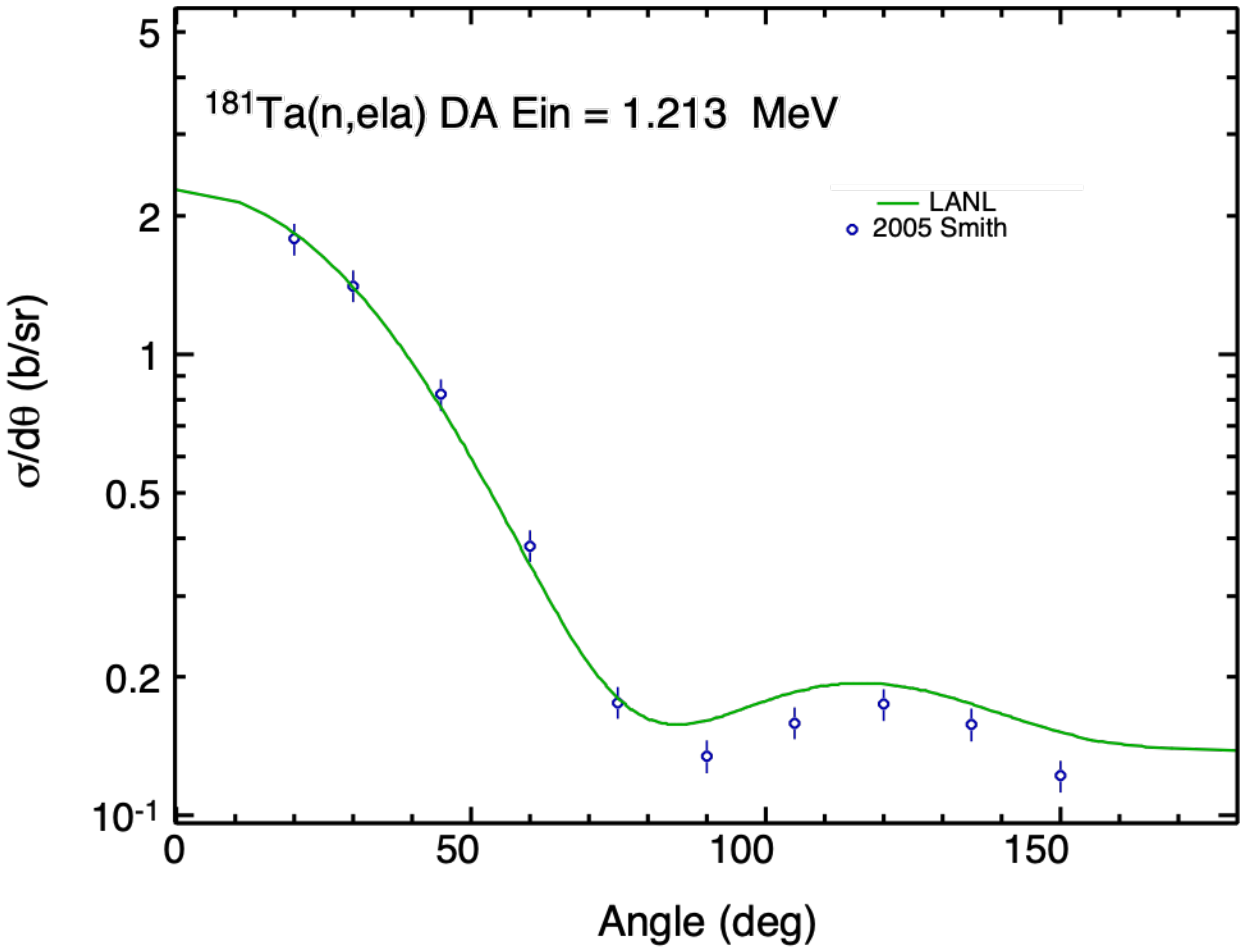

^{181}Ta(n,ela) DA Ein = 0.911 MeV
LANL
2005 Smith
σ/dθ (b/sr)
2
1
0.5
0.2
0
50
100
150
Angle (deg)

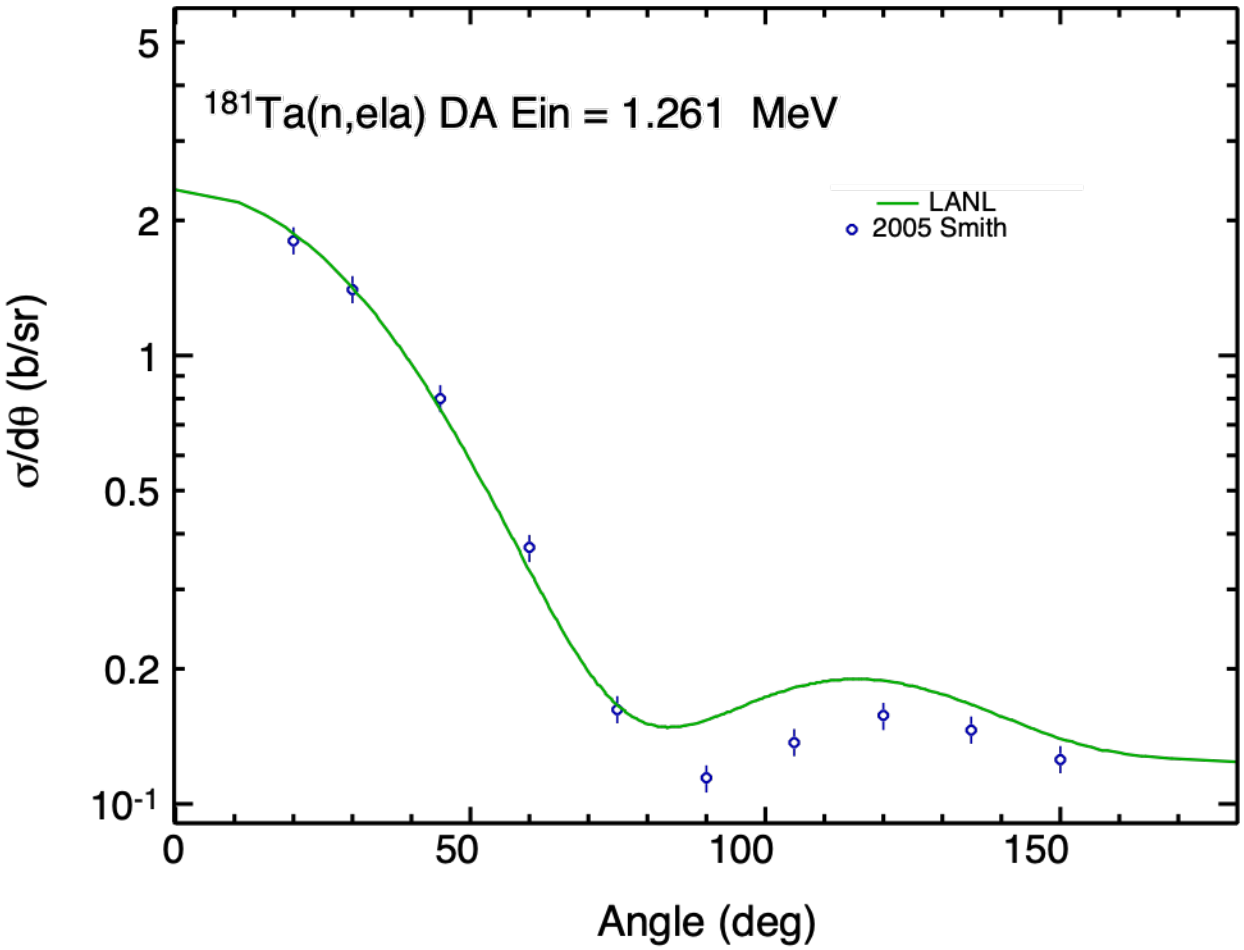

^{181}Ta(n,ela) DA Ein = 0.967 MeV
LANL
2005 Smith
σ/dθ (b/sr)
2
1
0.5
0.2
0
50
100
150
Angle (deg)

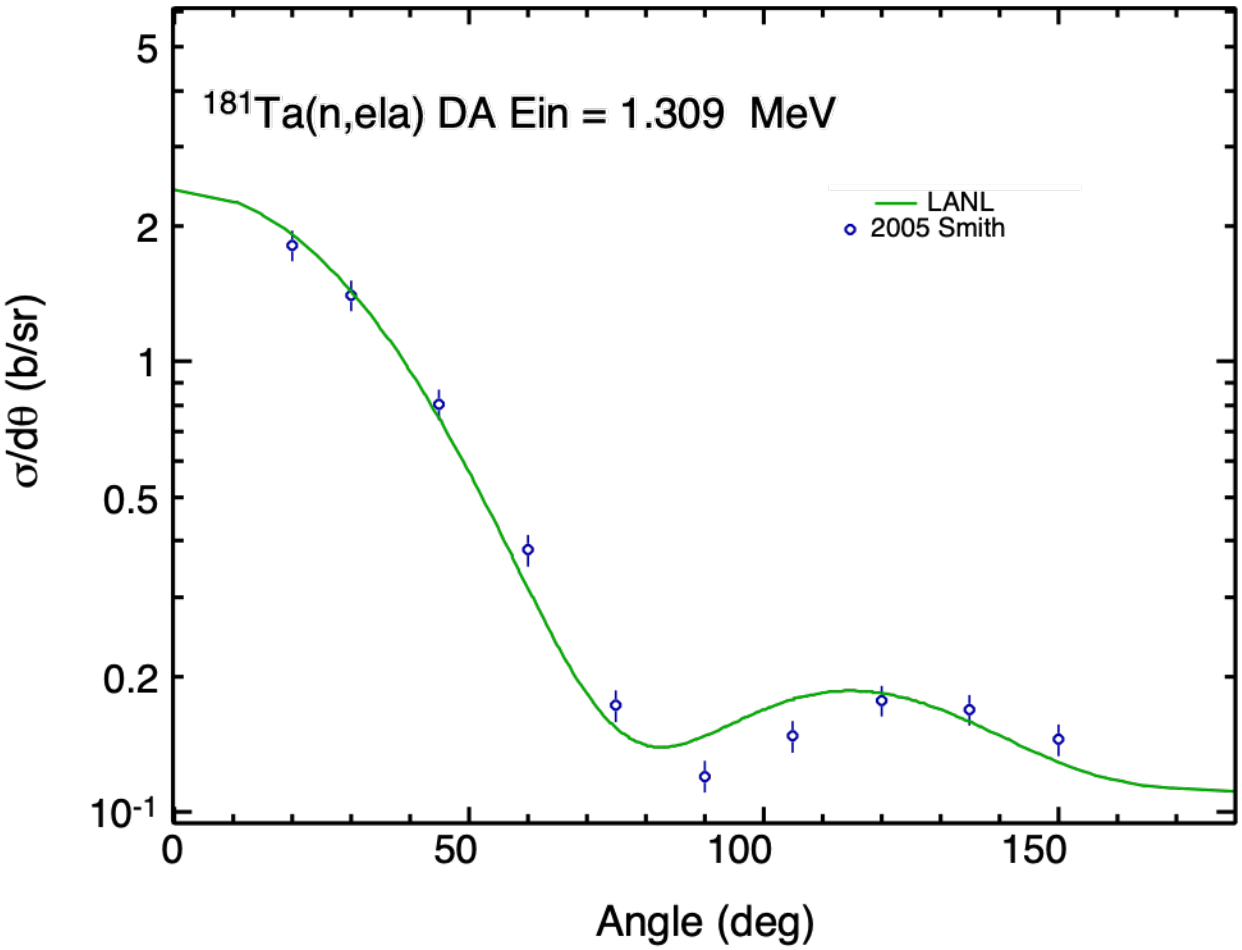

181Ta(n,ela) DA Ein = 1.015 MeV
LANL
2005 Smith
σ/dθ (b/sr)
2
1
0.5
0.2
0
50
100
150
Angle (deg)

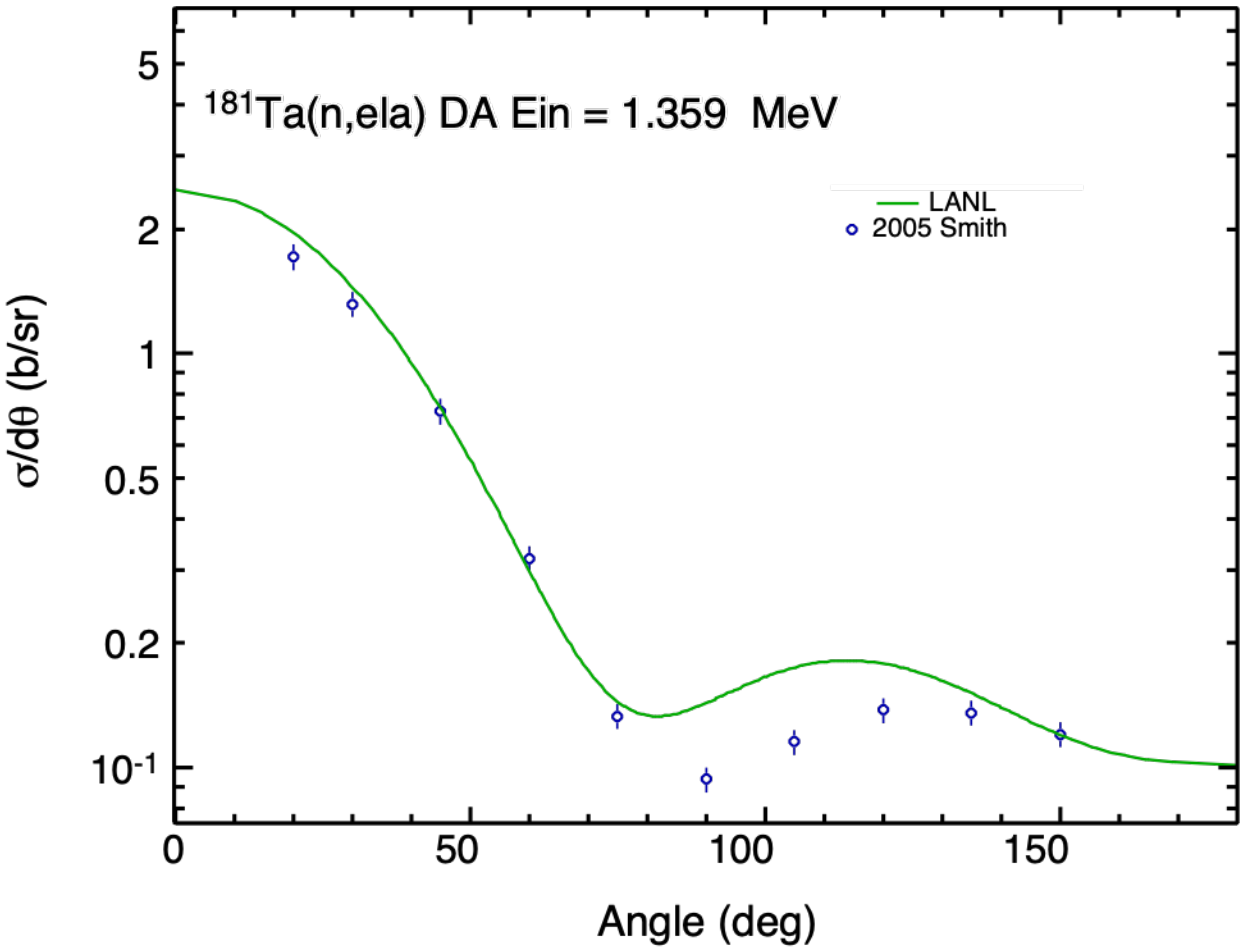

^{181}Ta(n,ela) DA Ein = 1.110 MeV
LANL
2005 Smith
σ/dθ (b/sr)
Angle (deg)
5
2
1
0.5
0.2
0
50
100
150

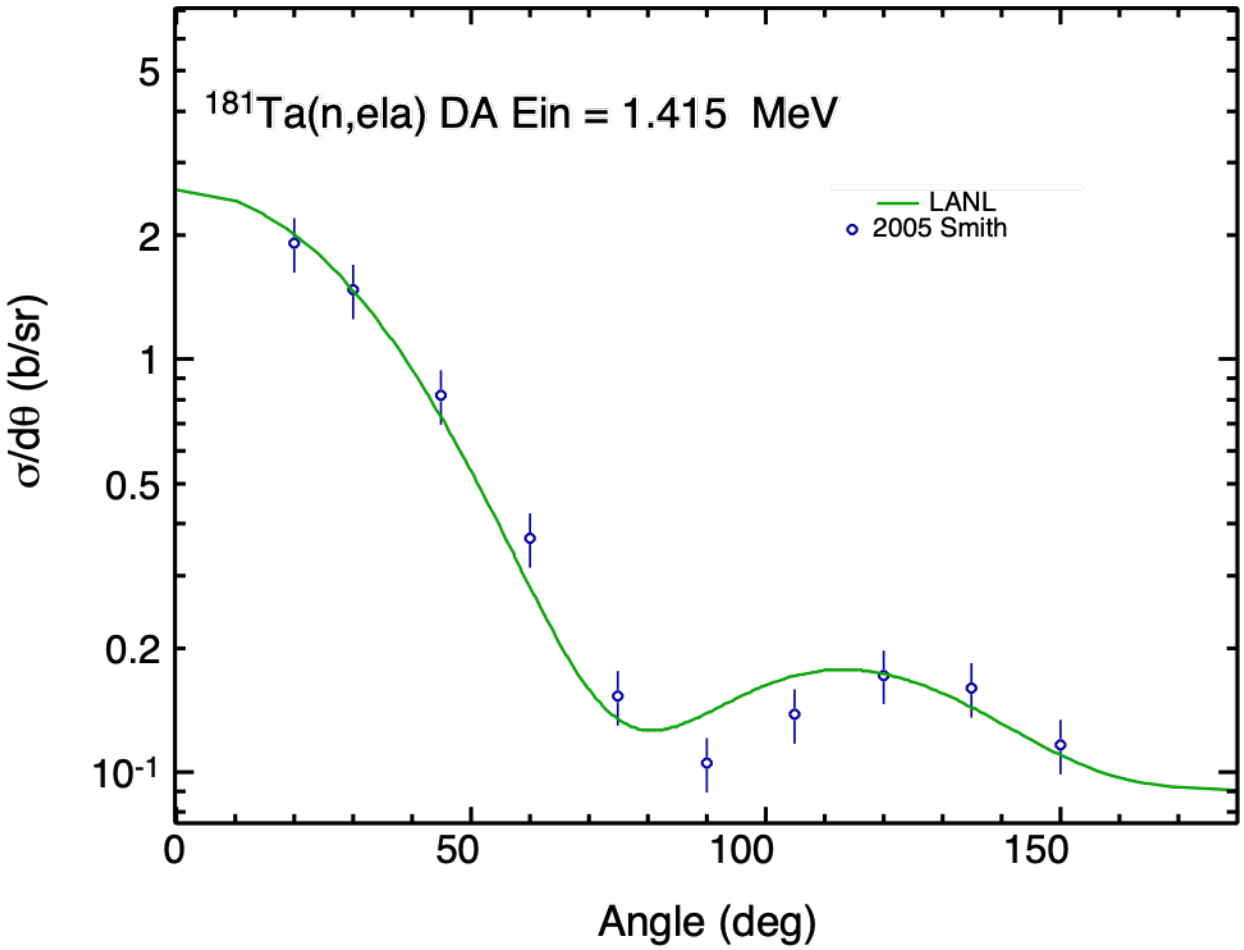

^{181}Ta(n,ela) DA Ein = 1.163 MeV
LANL
2005 Smith
σ/dθ (b/sr)
Angle (deg)
5
2
1
0.5
0.2
0
50
100
150

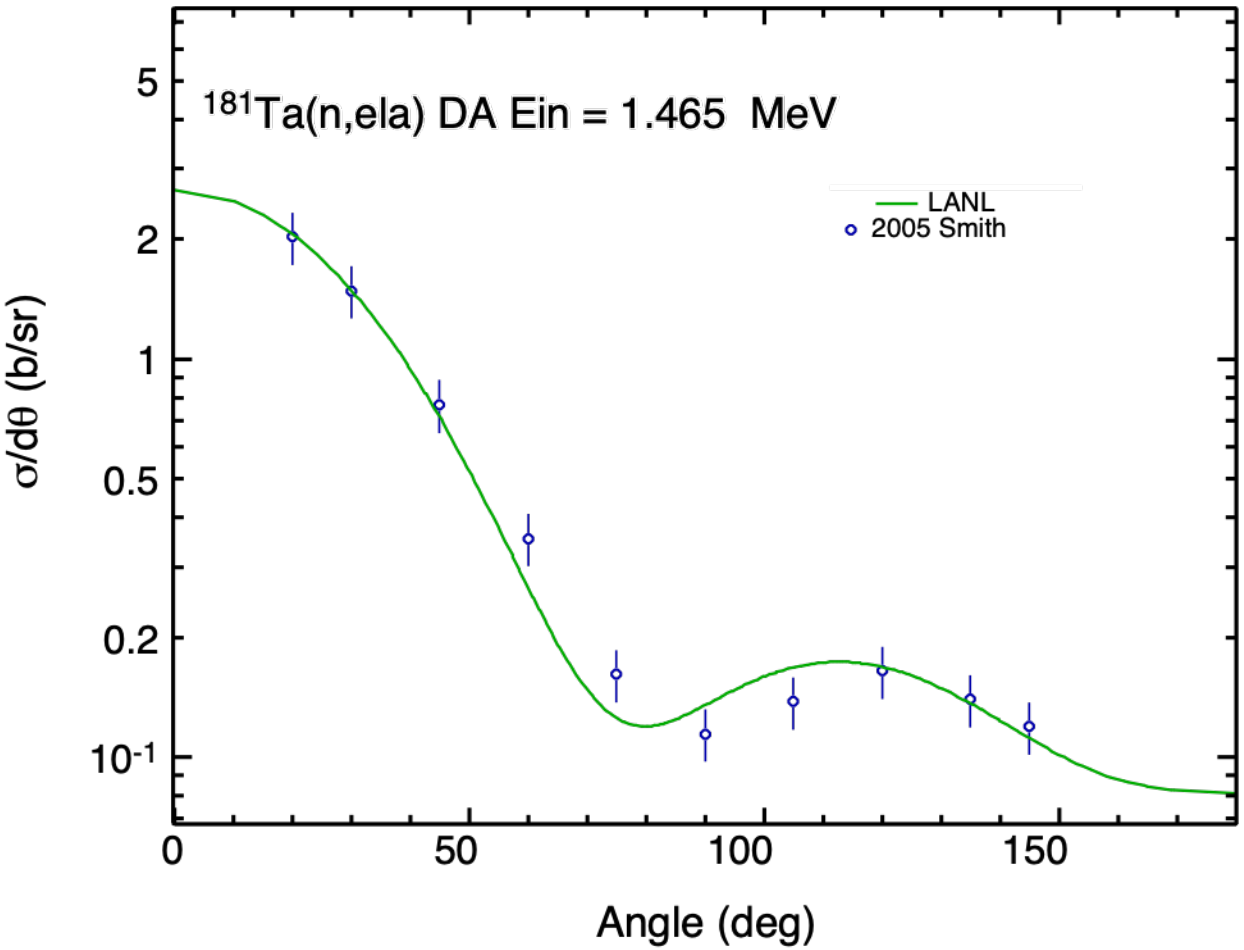

181Ta(n,ela) DA Ein = 1.213 MeV
LANL
2005 Smith
σ/dθ (b/sr)
Angle (deg)
5
2
1
0.5
0.2
10-1
0
50
100
150


^{181}Ta(n,ela) DA Ein = 1.261 MeV
LANL
2005 Smith
σ/dθ (b/sr)
5
2
1
0.5
0.2
10^{-1}
0
50
100
150
Angle (deg)

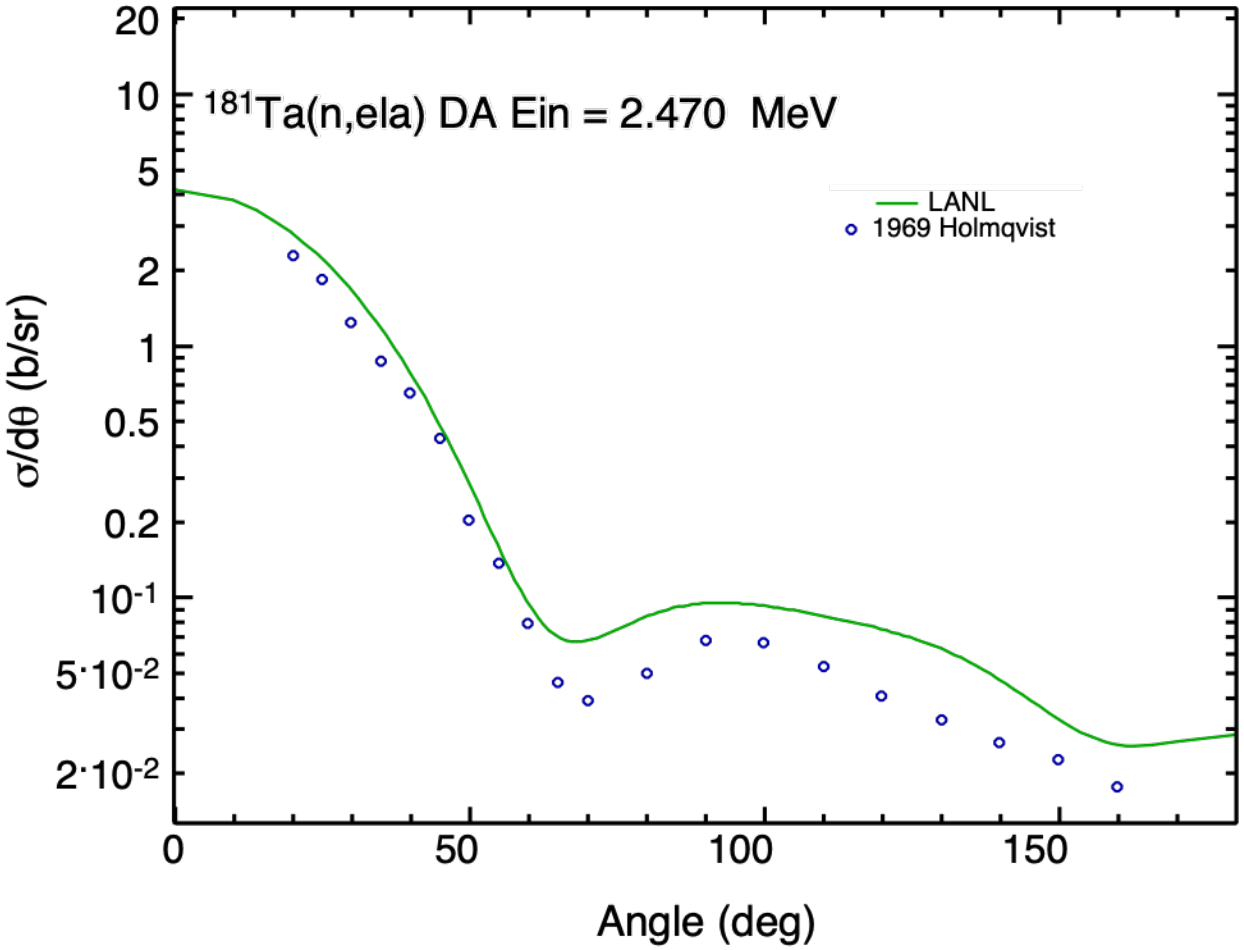

181Ta(n,ela) DA Ein = 1.309 MeV
LANL
2005 Smith
σ/dθ (b/sr)
5
2
1
0.5
0.2
10-1
0
50
100
150
Angle (deg)

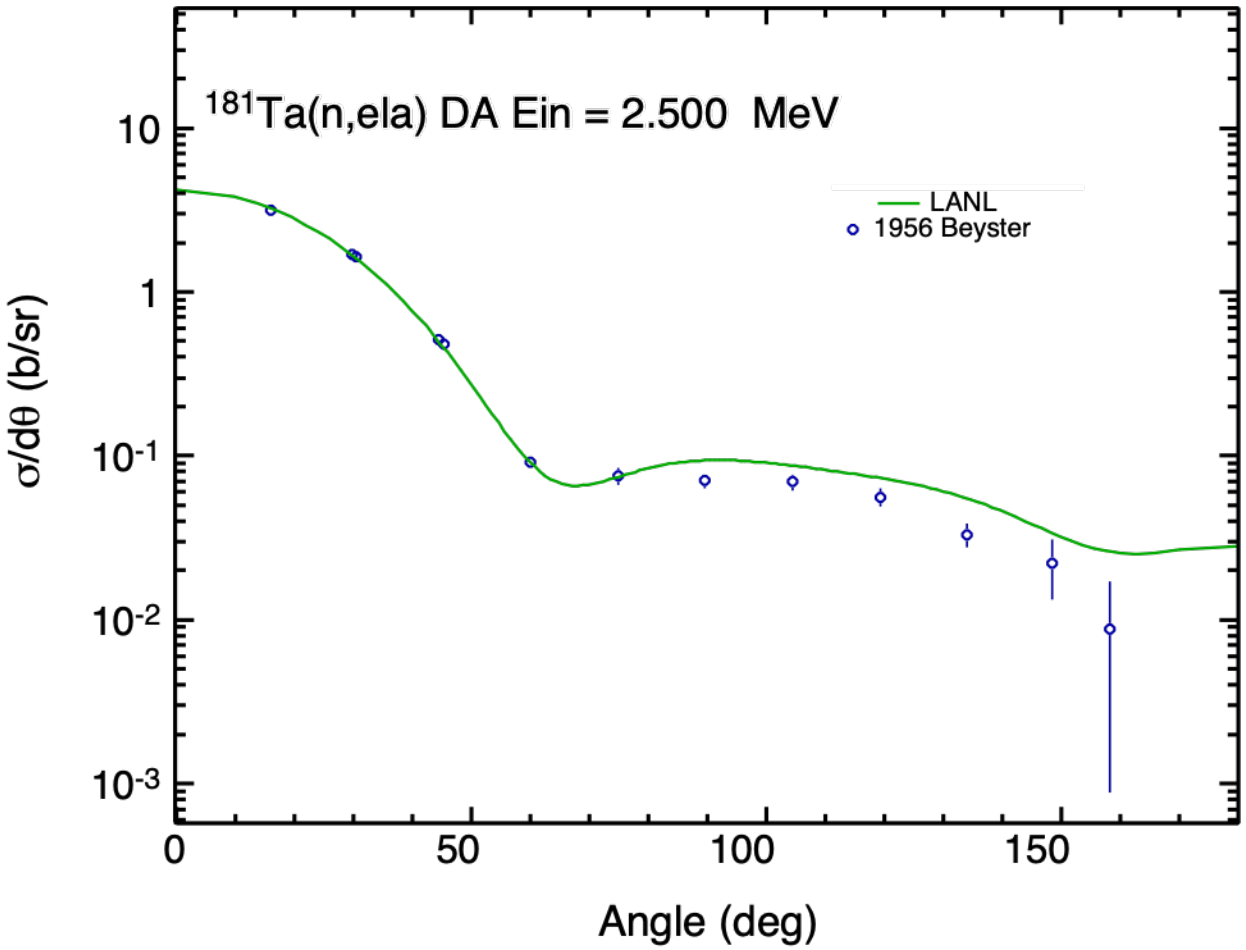

181Ta(n,ela) DA Ein = 1.359 MeV
LANL
2005 Smith
σ/dθ (b/sr)
Angle (deg)
5
2
1
0.5
0.2
10-1
0
50
100
150

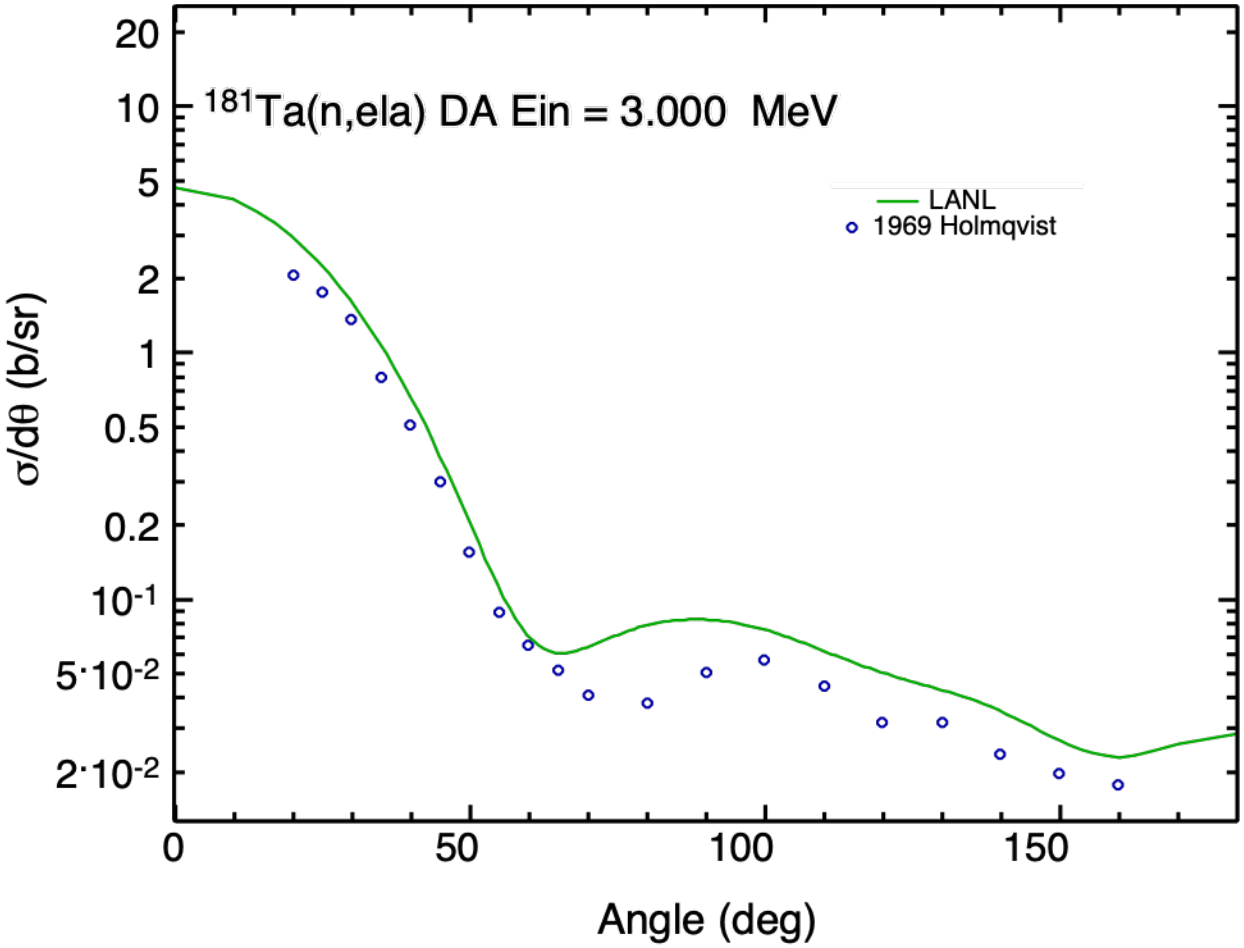

^{181}Ta(n,ela) DA Ein = 1.415 MeV
LANL
2005 Smith
σ/dθ (b/sr)
5
2
1
0.5
0.2
10^{-1}
0
50
100
150
Angle (deg)

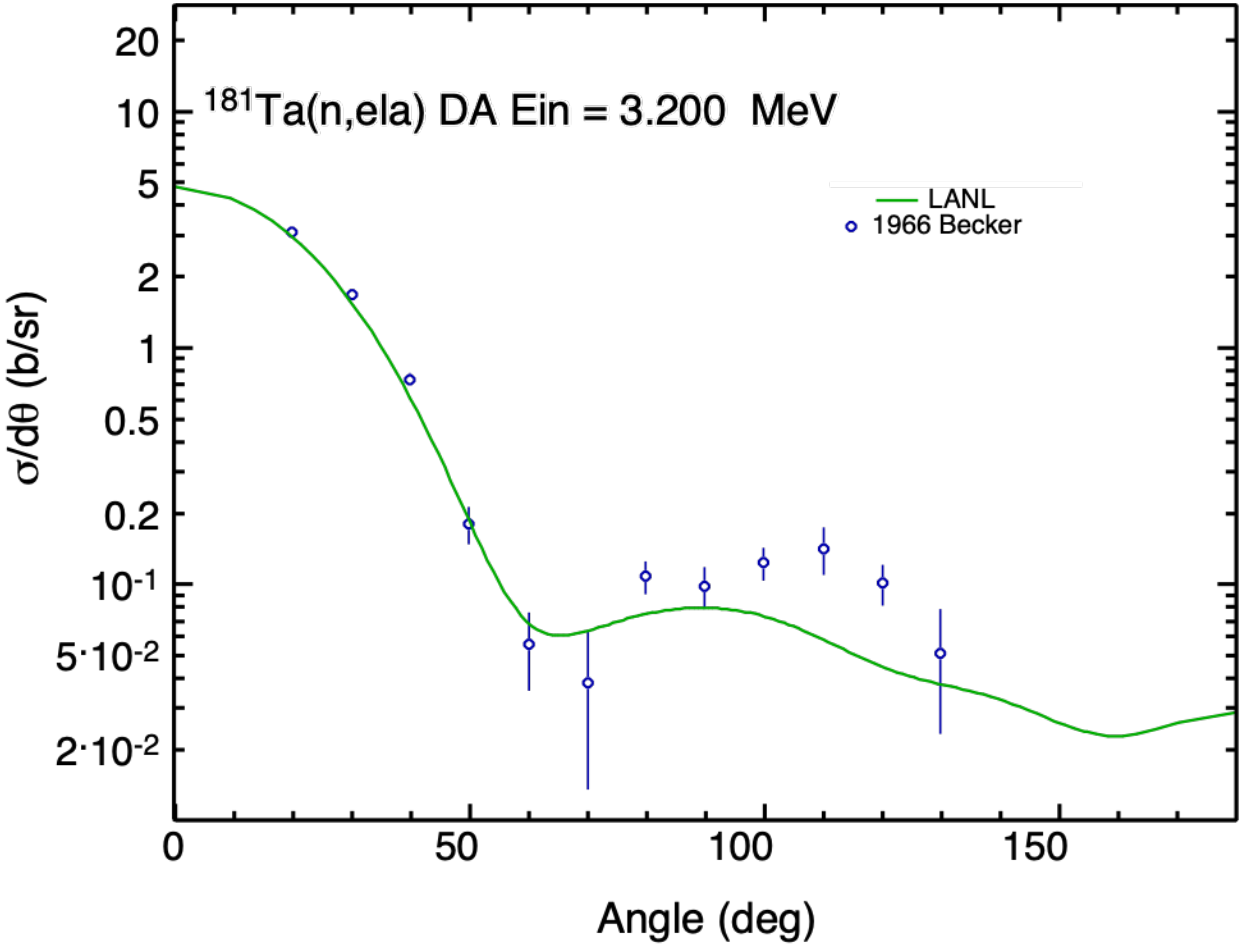

181Ta(n,ela) DA Ein = 1.465 MeV
LANL
2005 Smith
σ/dθ (b/sr)
5
2
1
0.5
0.2
10-1
0
50
100
150
Angle (deg)

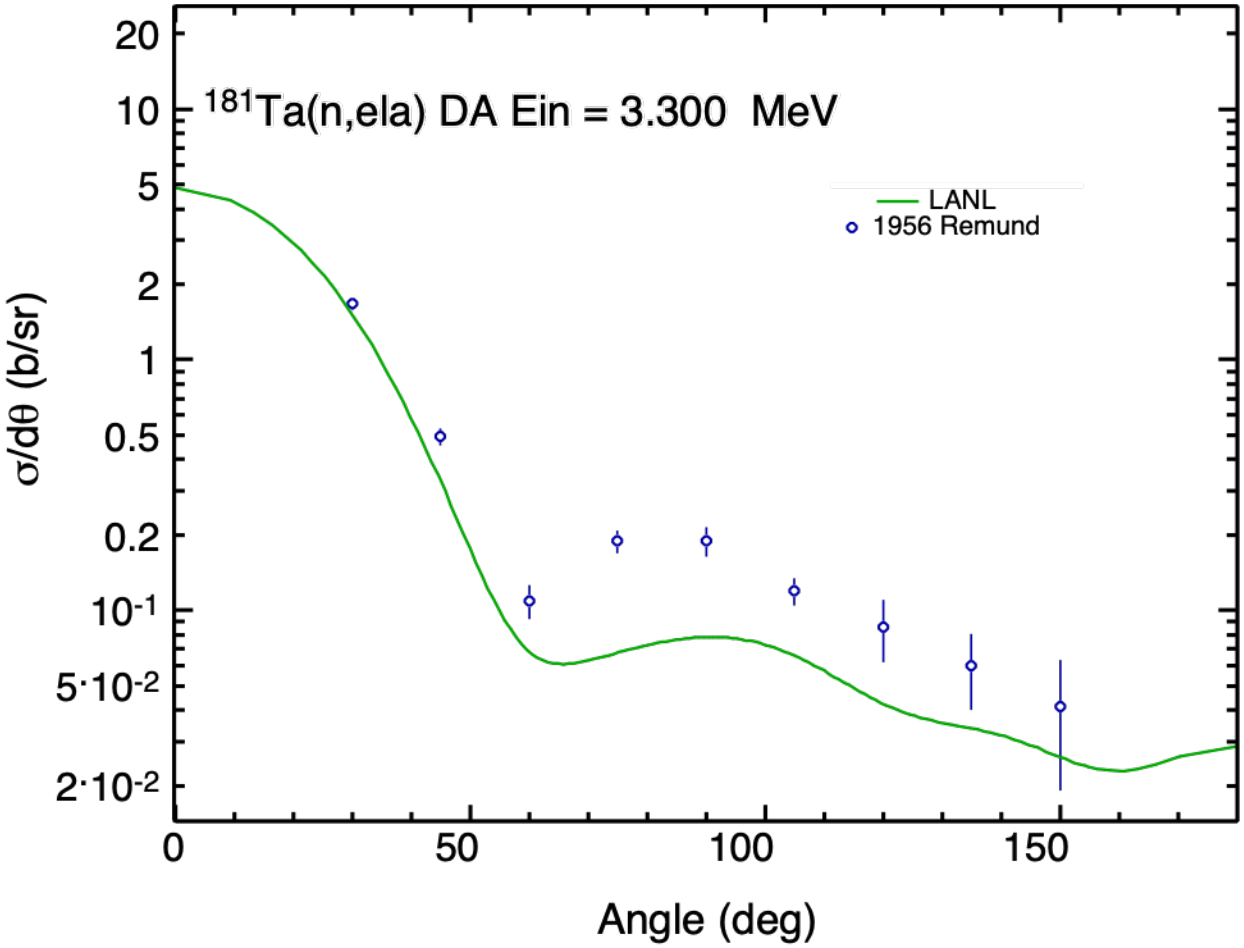
Various experiments: 2.5 - 15.2 MeV

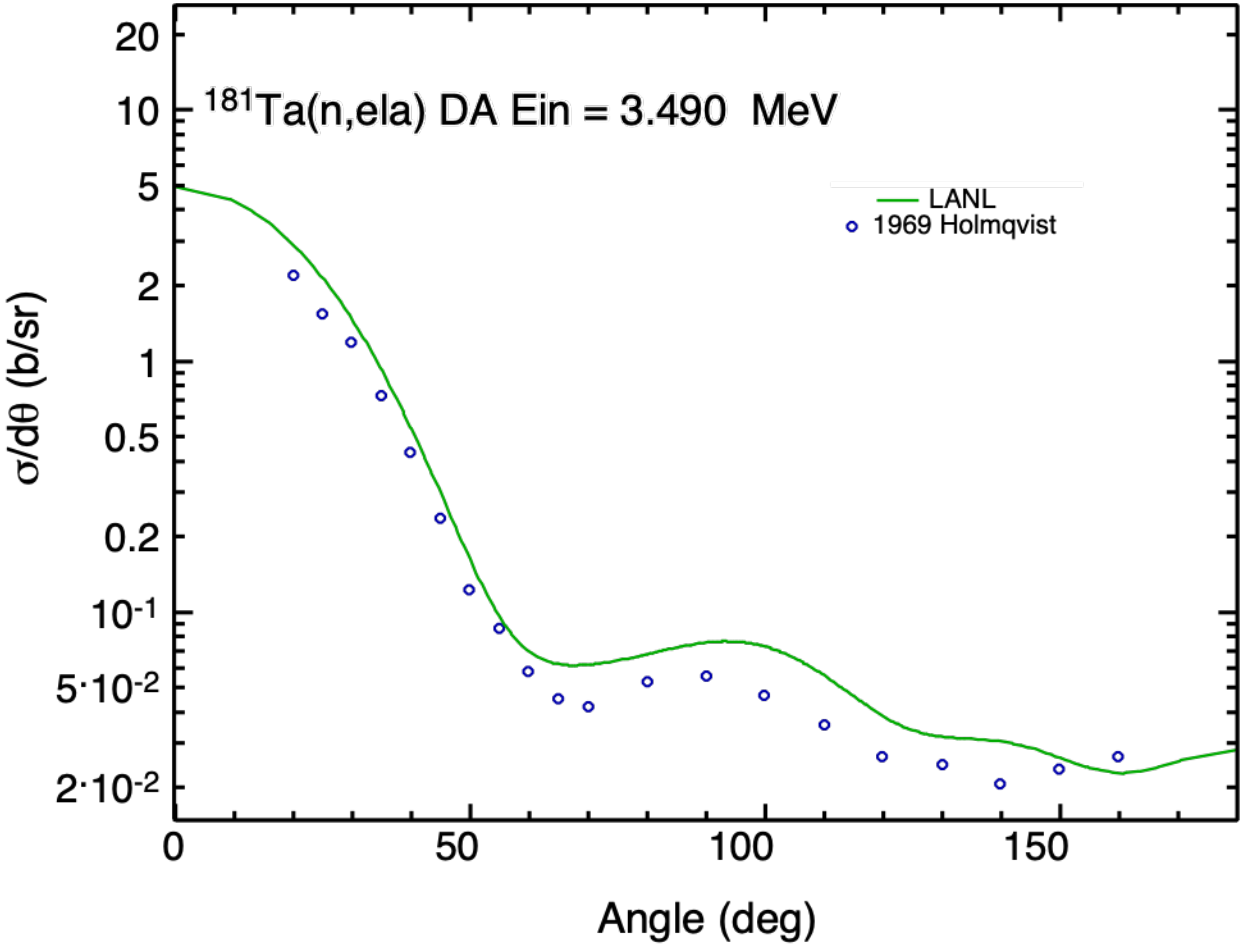


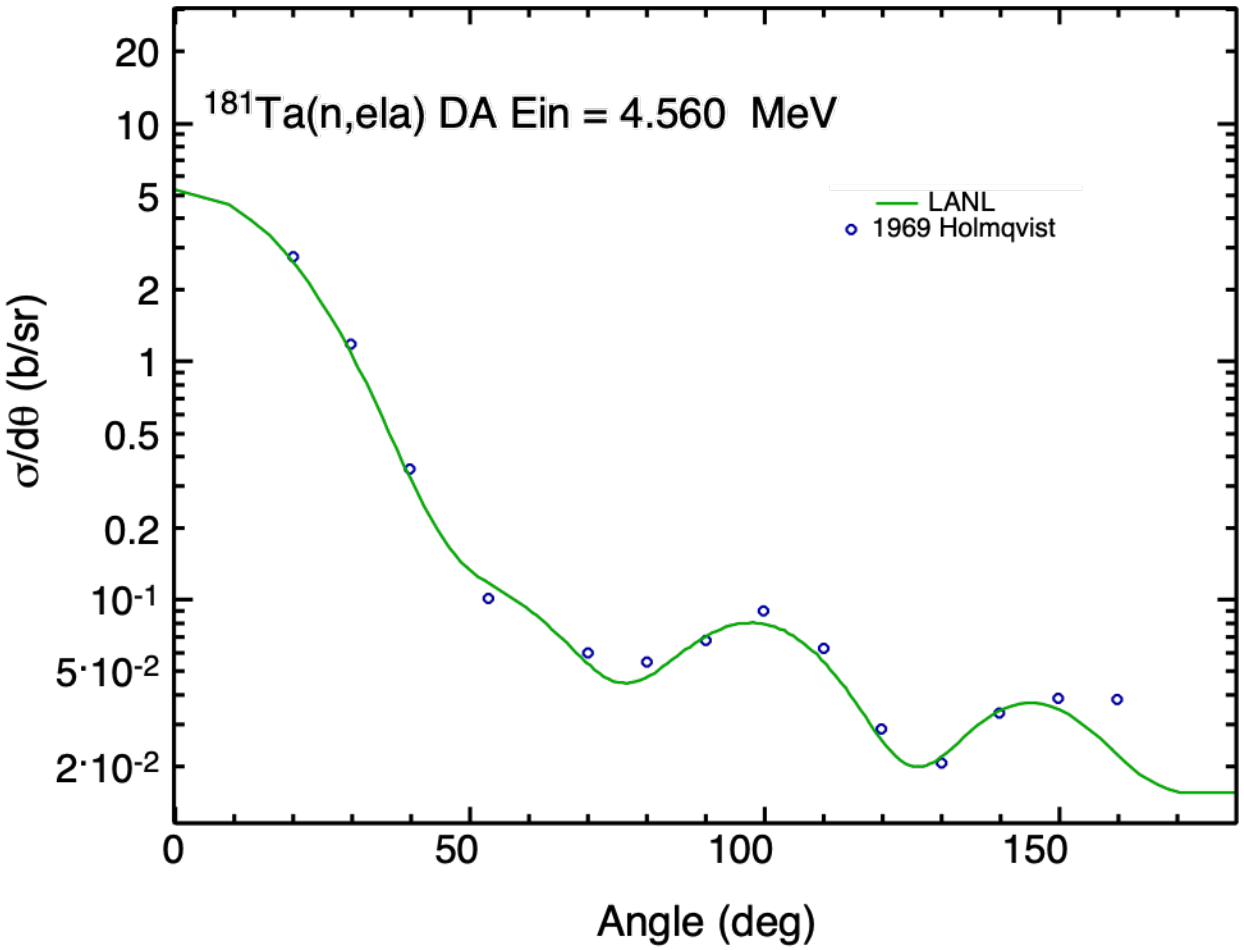

^{181}Ta(n,ela) DA Ein = 2.500 MeV
LANL
1956 Beyster
σ/dθ (b/sr)
10
1
10-1
10-2
10-3
0
50
100
150
Angle (deg)

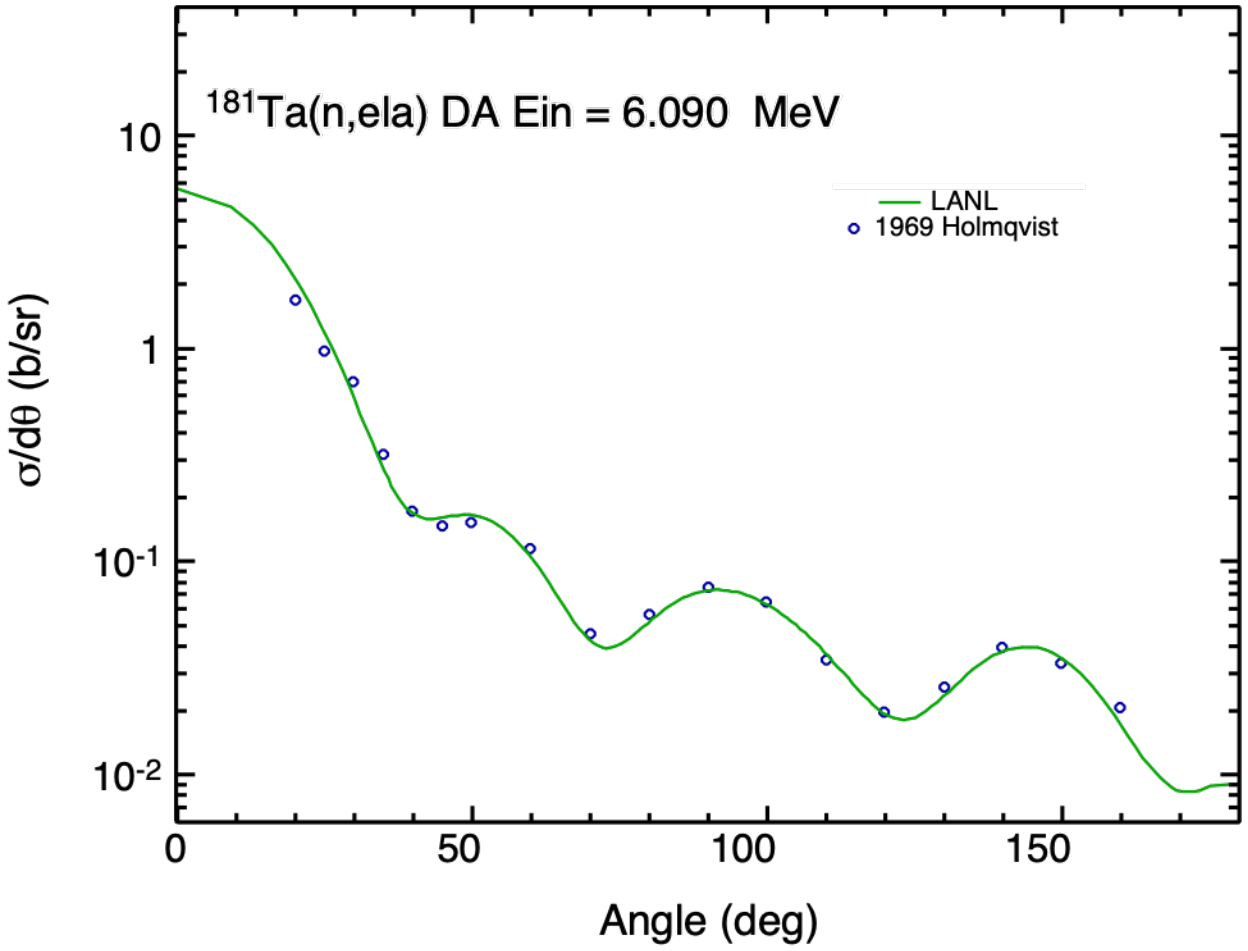

^{181}Ta(n,ela) DA Ein = 3.000 MeV
LANL
1969 Holmqvist
σ/dθ (b/sr)
20
10
5
2
1
0.5
0.2
10^{-1}
5·10^{-2}
2·10^{-2}
0
50
100
150
Angle (deg)

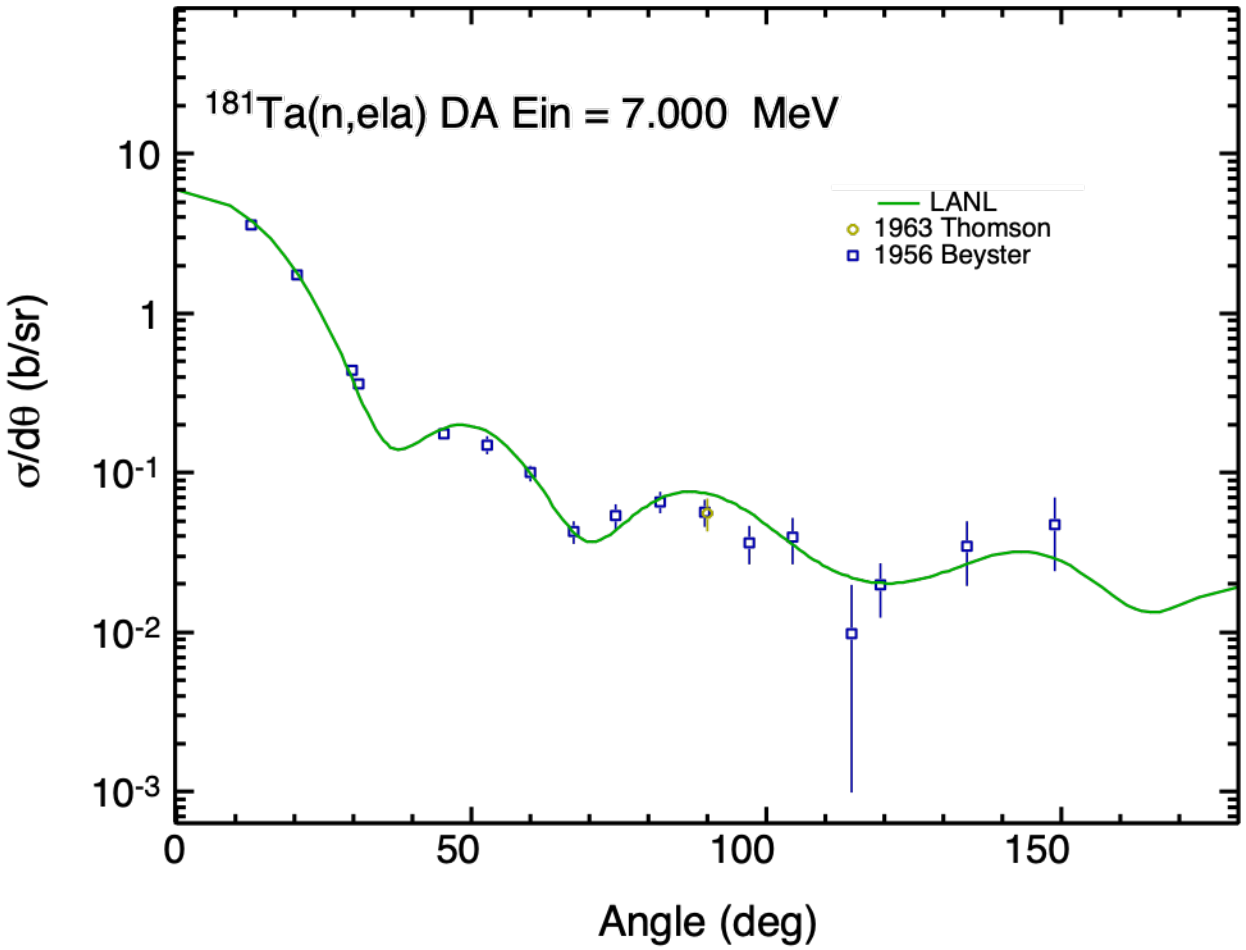

181Ta(n,ela) DA Ein = 3.200 MeV
LANL
1966 Becker
σ/dθ (b/sr)
Angle (deg)
20
10
5
2
1
0.5
0.2
10-1
5·10-2
2·10-2
0
50
100
150

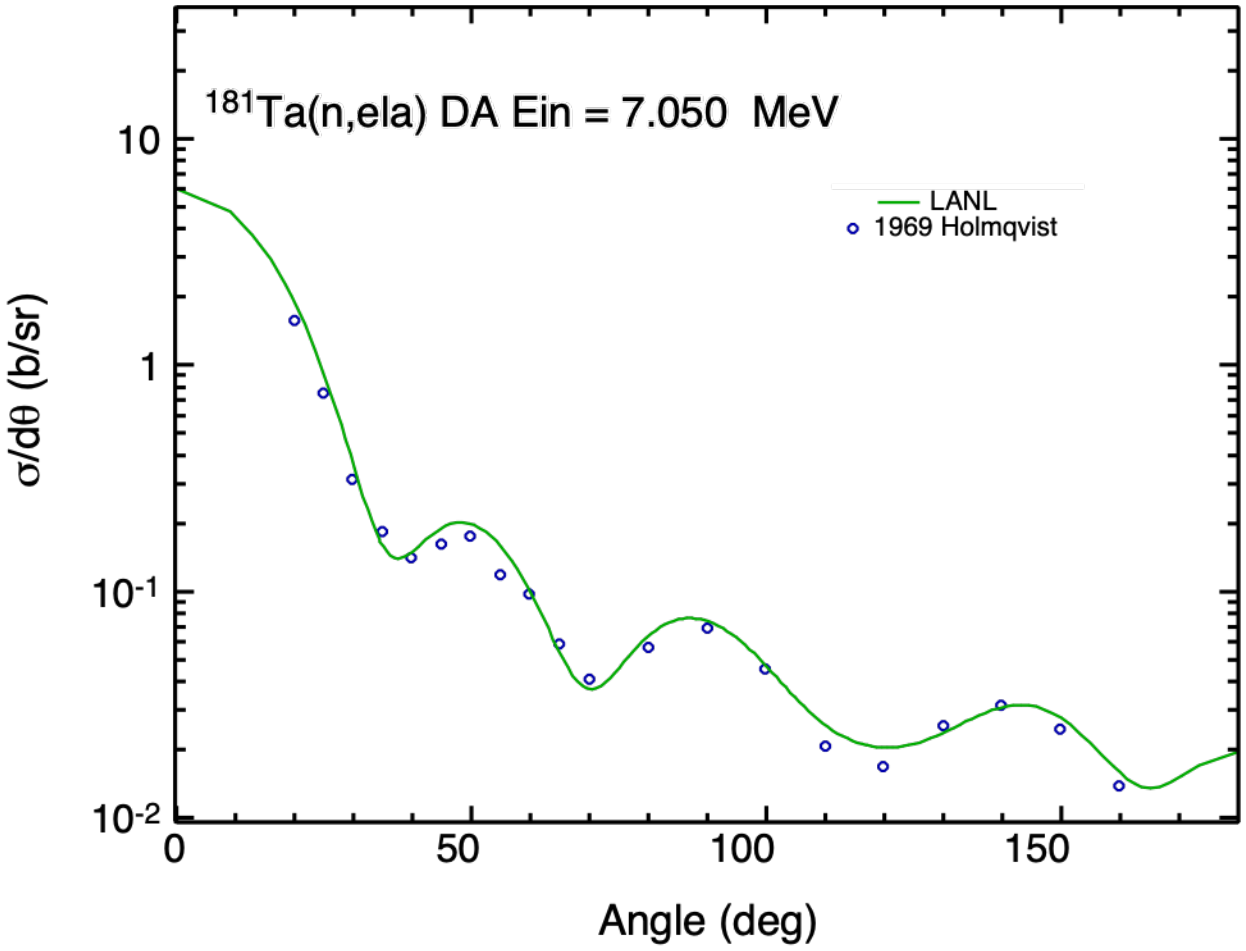

^{181}Ta(n,ela) DA Ein = 3.300 MeV
LANL
1956 Remund
σ/dθ (b/sr)
20
10
5
2
1
0.5
0.2
10-1
5·10-2
2·10-2
0
50
100
150
Angle (deg)

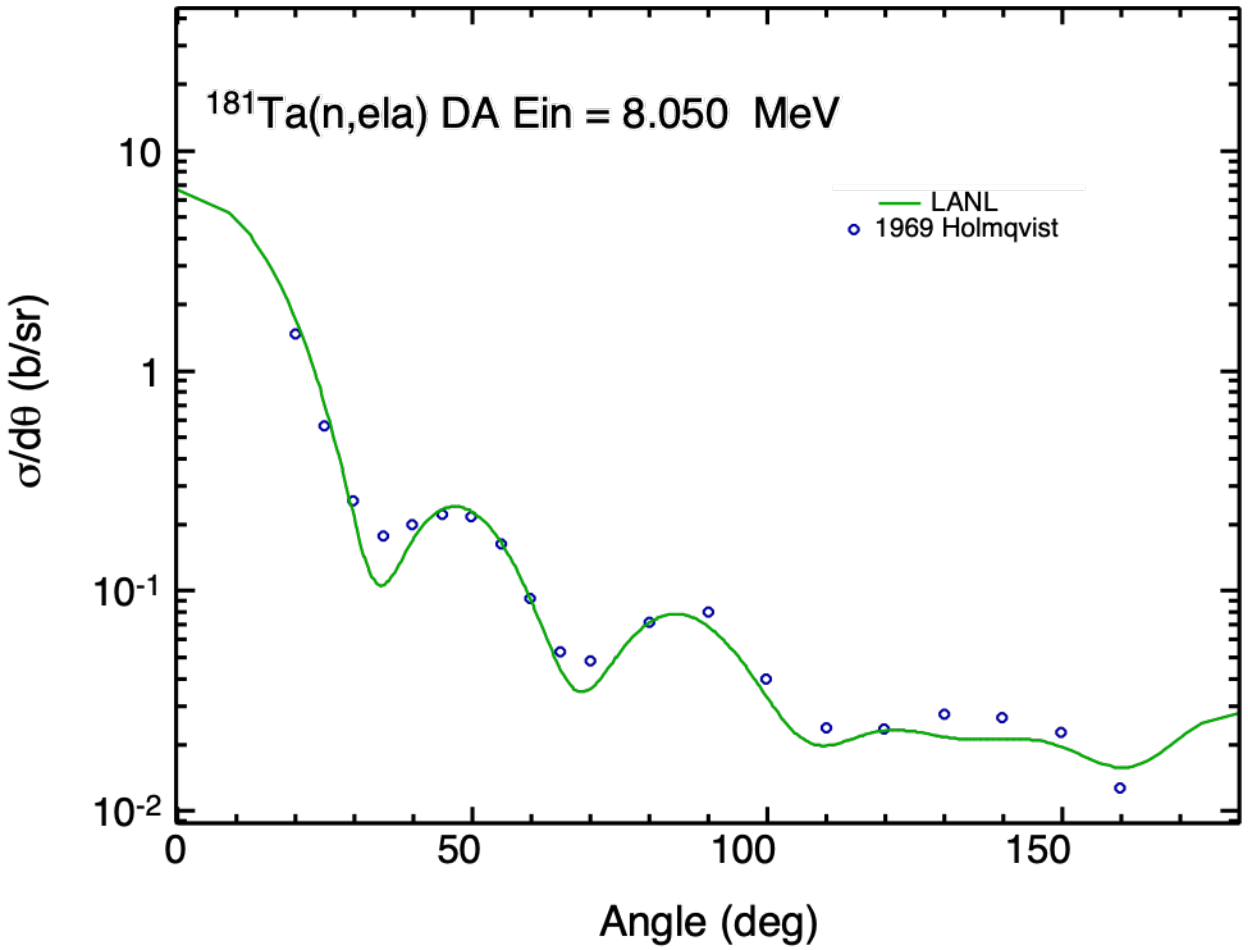

^{181}Ta(n,ela) DA Ein = 3.490 MeV
LANL
1969 Holmqvist
σ/dθ (b/sr)
Angle (deg)
20
10
5
2
1
0.5
0.2
10-1
5·10-2
2·10-2
0
50
100
150

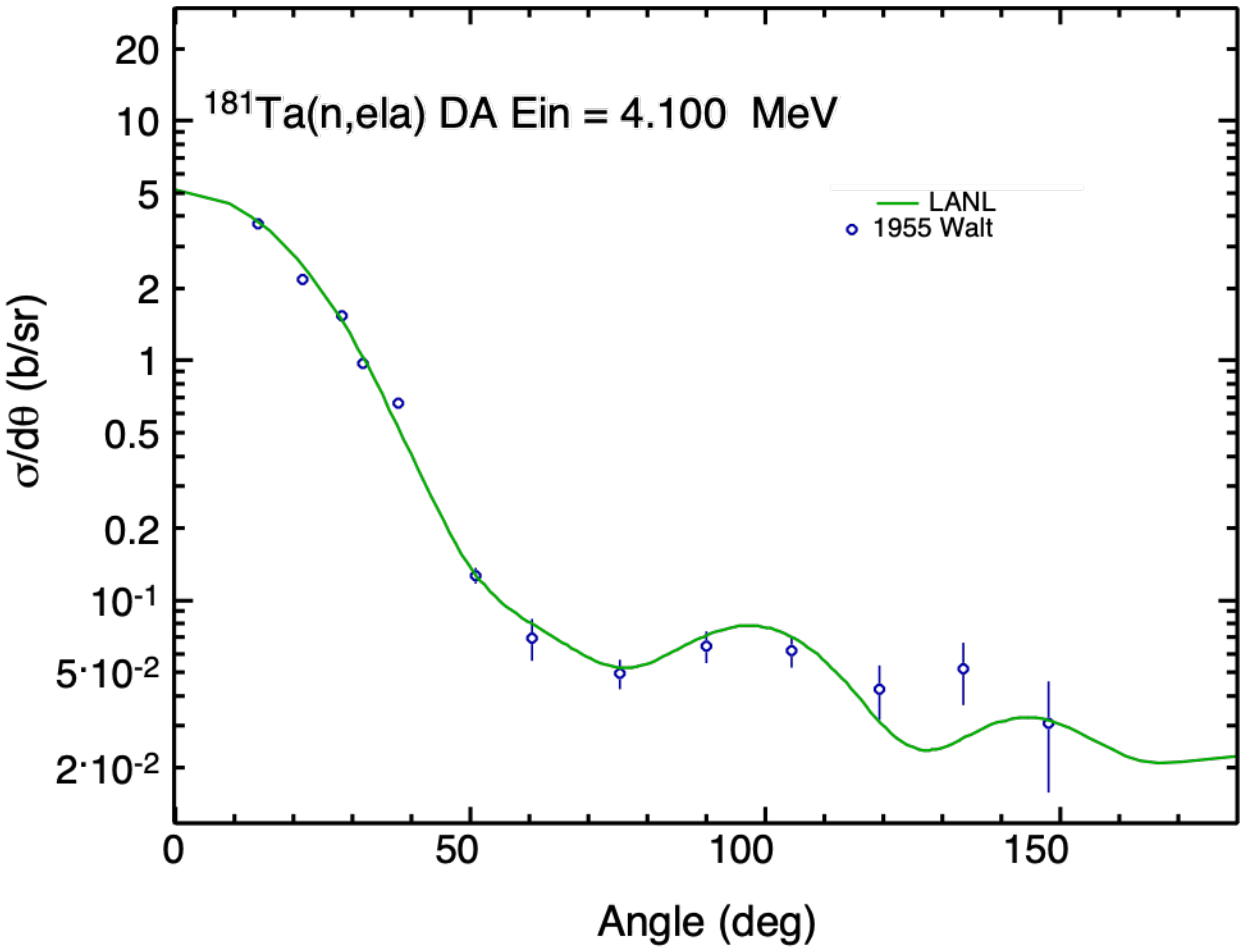

181Ta(n,ela) DA Ein = 4.560 MeV
LANL
1969 Holmqvist
σ/dθ (b/sr)
Angle (deg)
20
10
5
2
1
0.5
0.2
10-1
5·10-2
2·10-2
0
50
100
150

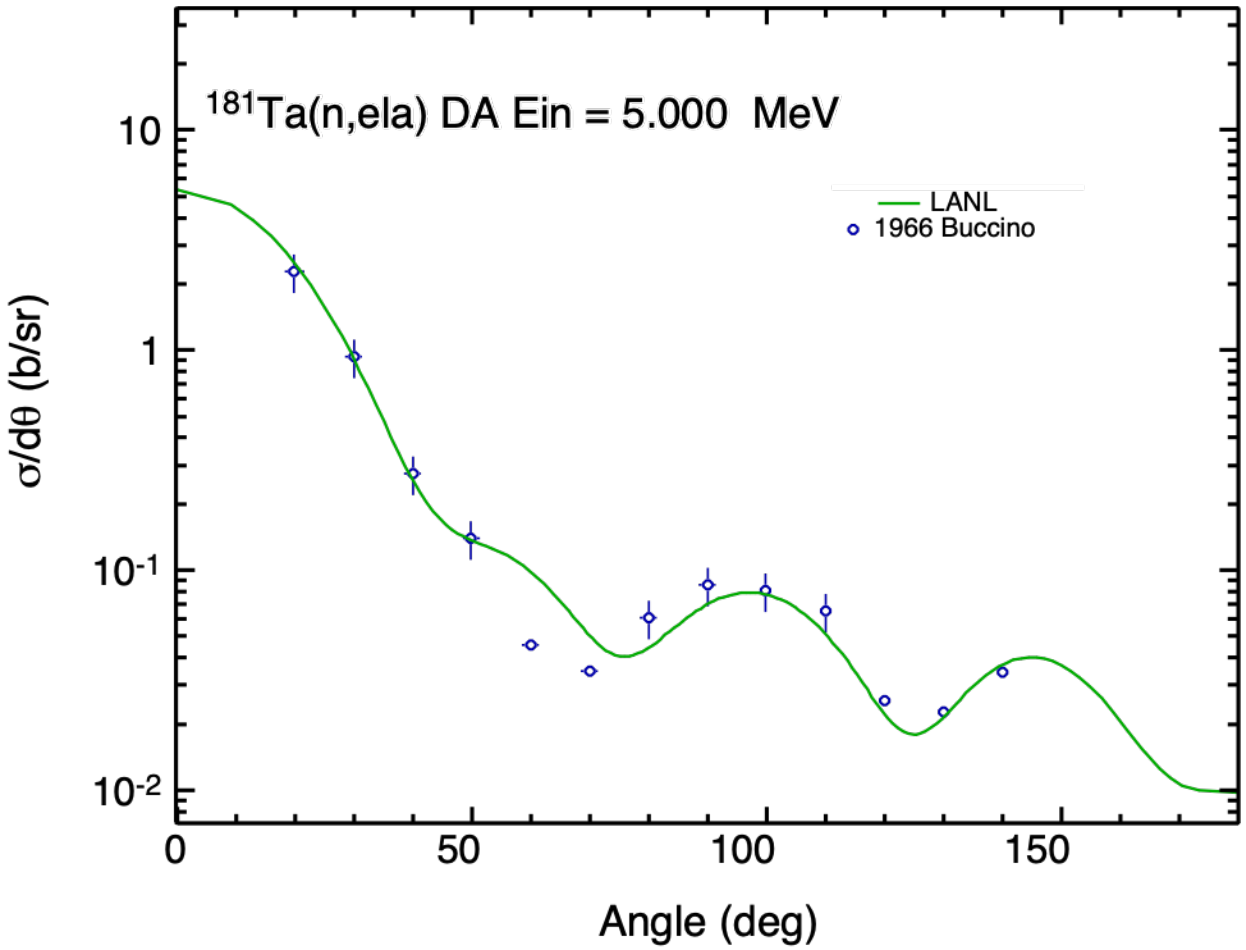

^{181}Ta(n,ela) DA Ein = 6.090 MeV
LANL
1969 Holmqvist
σ/dθ (b/sr)
10
1
10-1
10-2
0
50
100
150
Angle (deg)

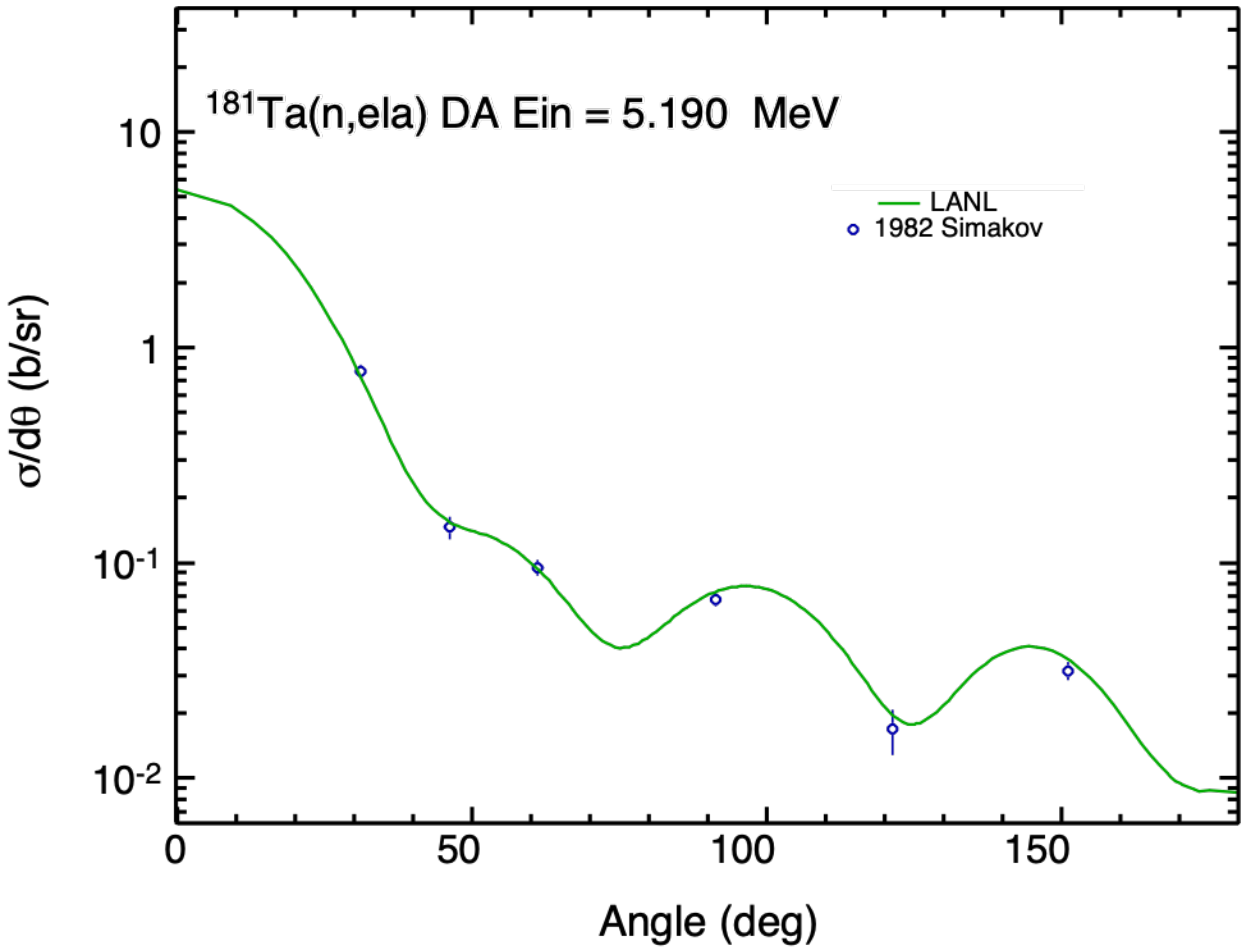

^{181}Ta(n,ela) DA Ein = 7.000 MeV
LANL
1963 Thomson
1956 Beyster
σ/dθ (b/sr)
10
1
10-1
10-2
10-3
0
50
100
150
Angle (deg)

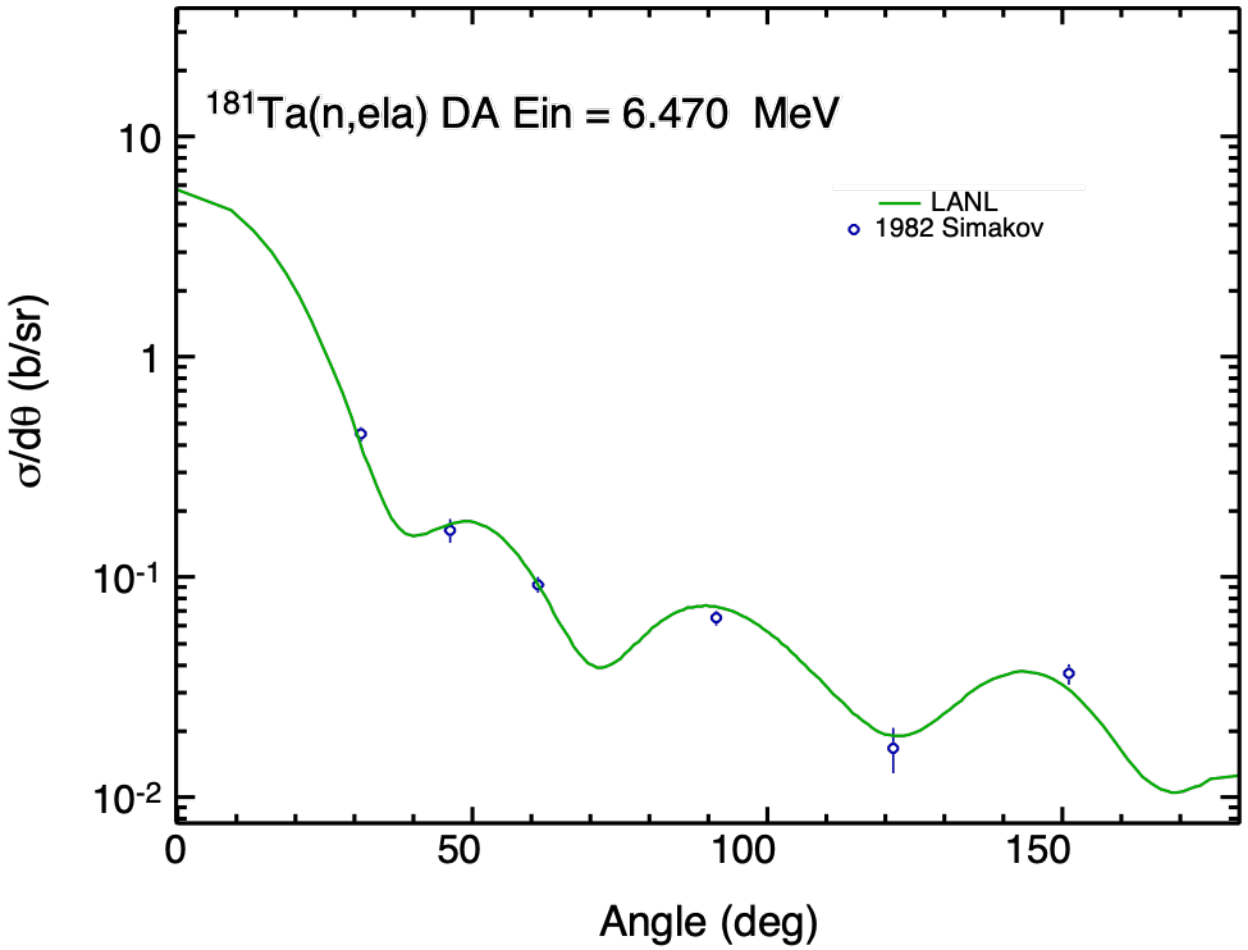

^{181}Ta(n,ela) DA Ein = 7.050 MeV
LANL
1969 Holmqvist
σ/dθ (b/sr)
10
1
10-1
10-2
0
50
100
150
Angle (deg)

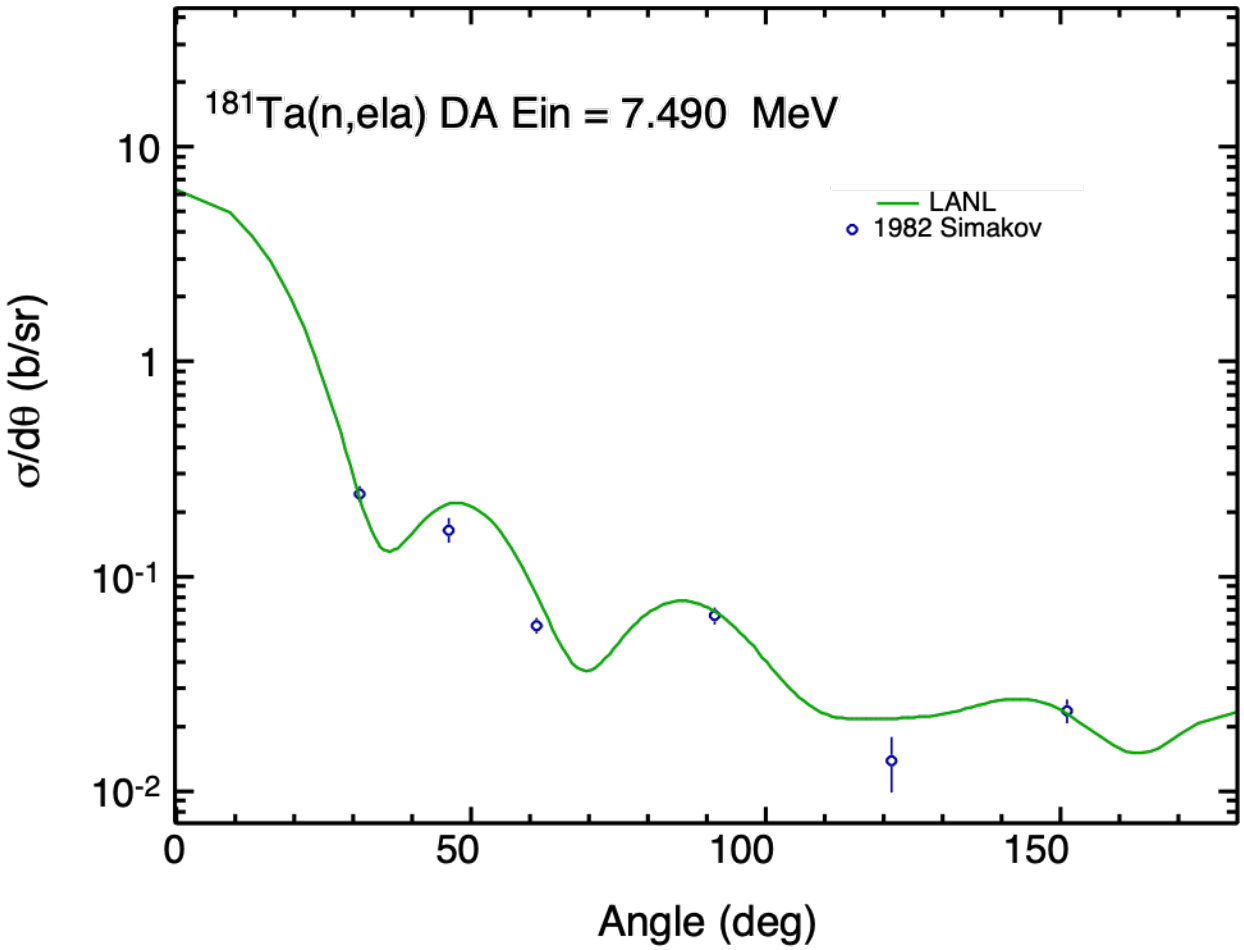

^{181}Ta(n,ela) DA Ein = 8.050 MeV
LANL
1969 Holmqvist
σ/dθ (b/sr)
10
1
10-1
10-2
0
50
100
150
Angle (deg)

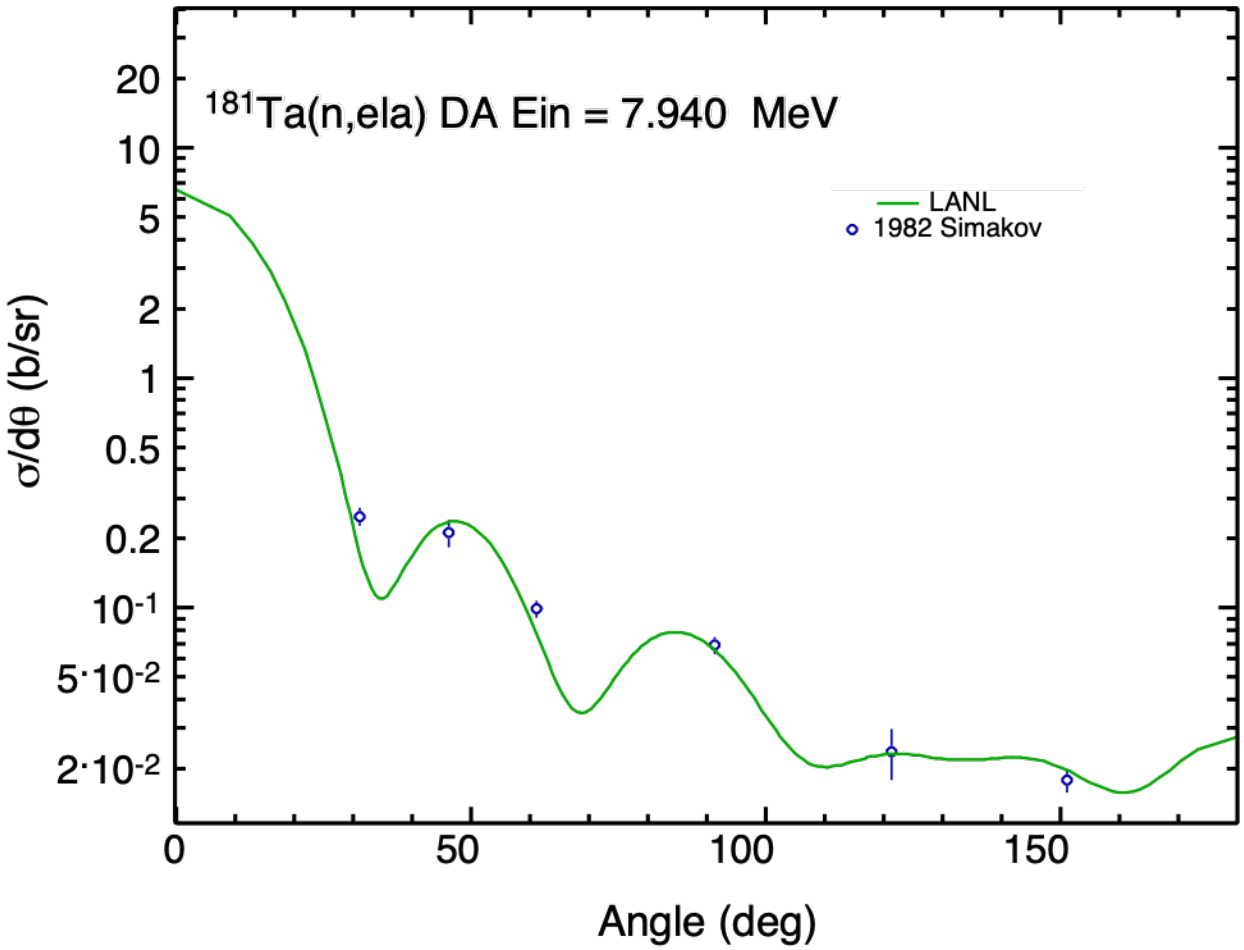

181Ta(n,ela) DA Ein = 4.100 MeV
LANL
1955 Walt
σ/dθ (b/sr)
20
10
5
2
1
0.5
0.2
10-1
5·10-2
2·10-2
0
50
100
150
Angle (deg)

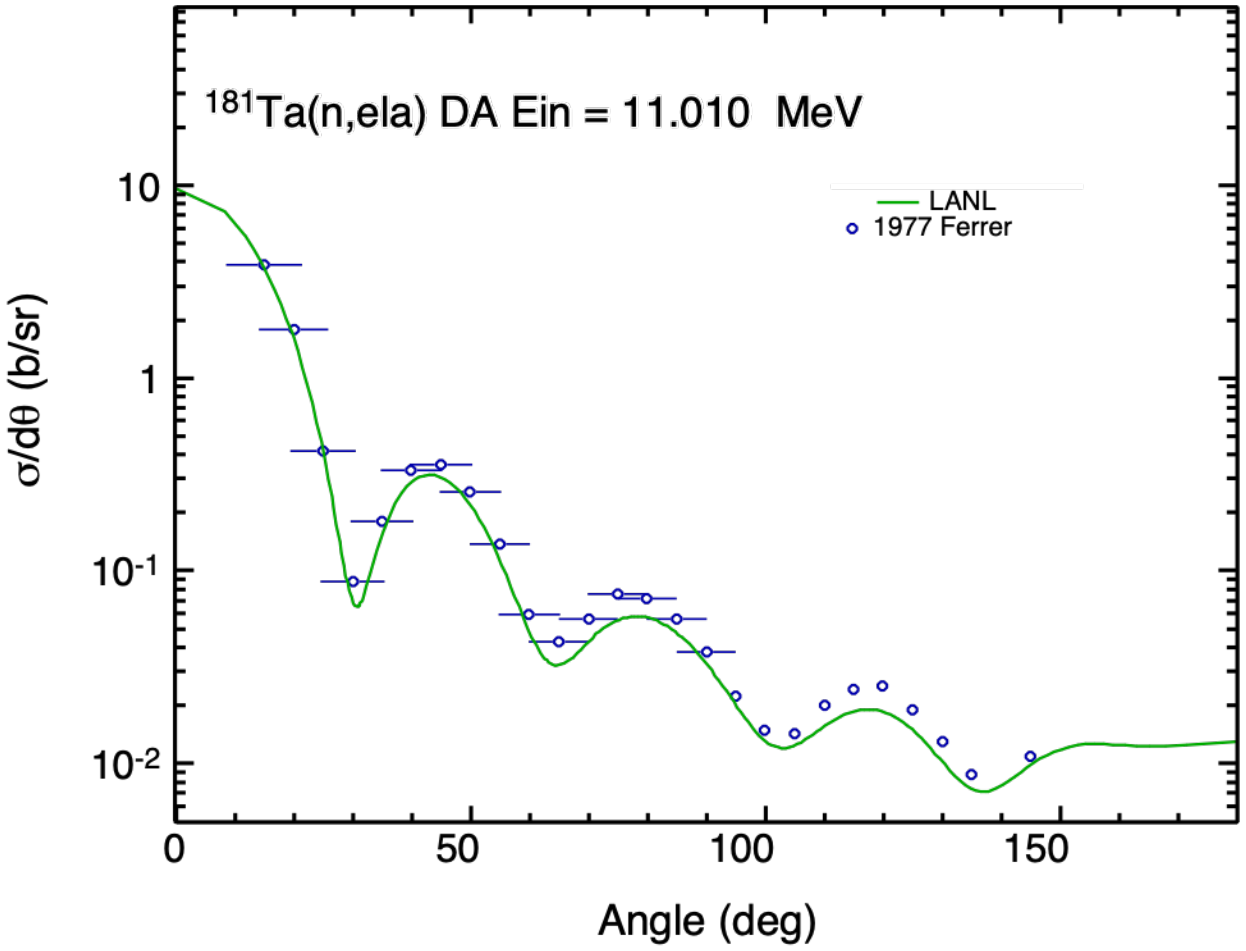

^{181}Ta(n,ela) DA Ein = 5.000 MeV
LANL
1966 Buccino
σ/dθ (b/sr)
10
1
10-1
10-2
0
50
100
150
Angle (deg)

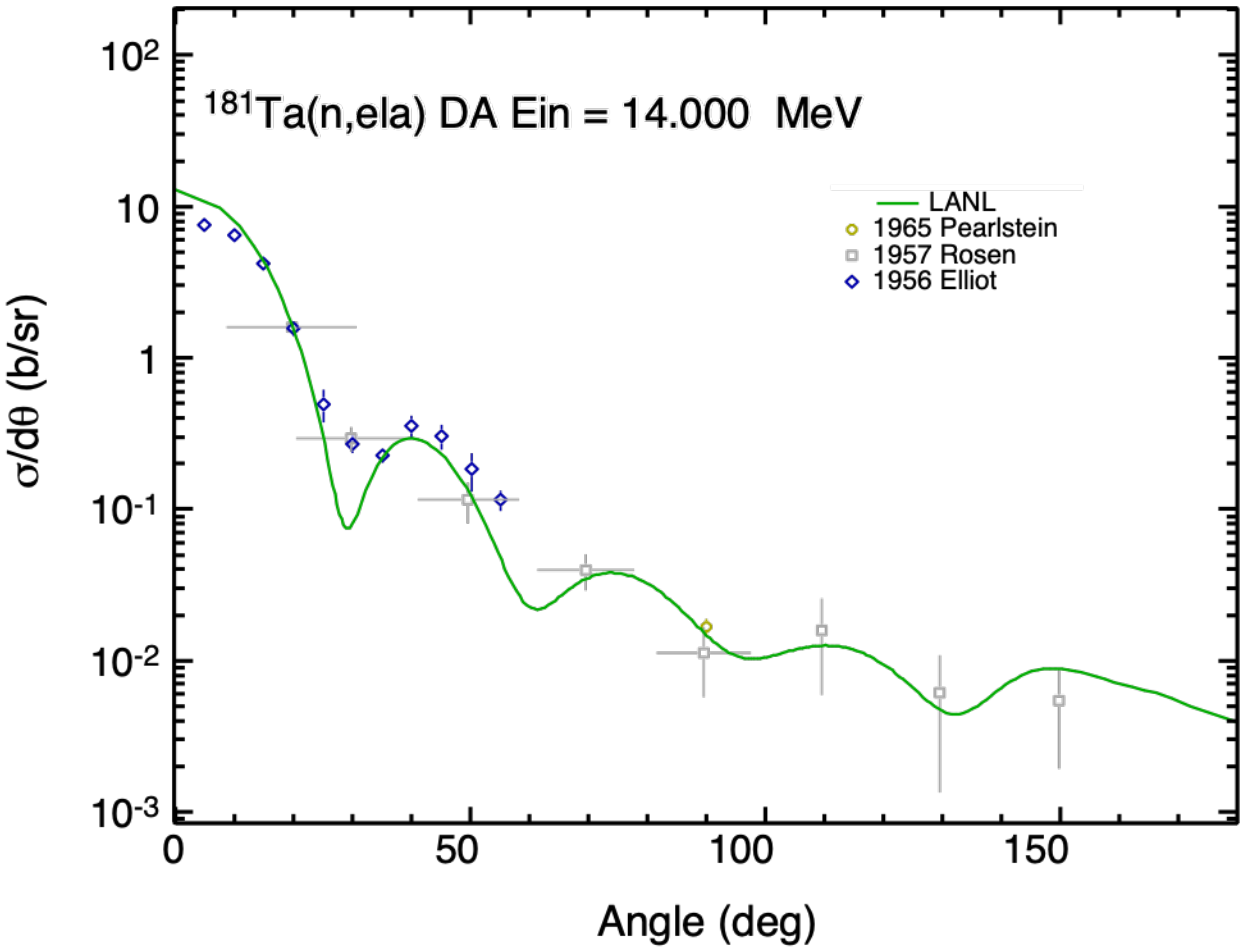

^{181}Ta(n,ela) DA Ein = 5.190 MeV
LANL
1982 Simakov
σ/dθ (b/sr)
10
1
10-1
10-2
0
50
100
150
Angle (deg)

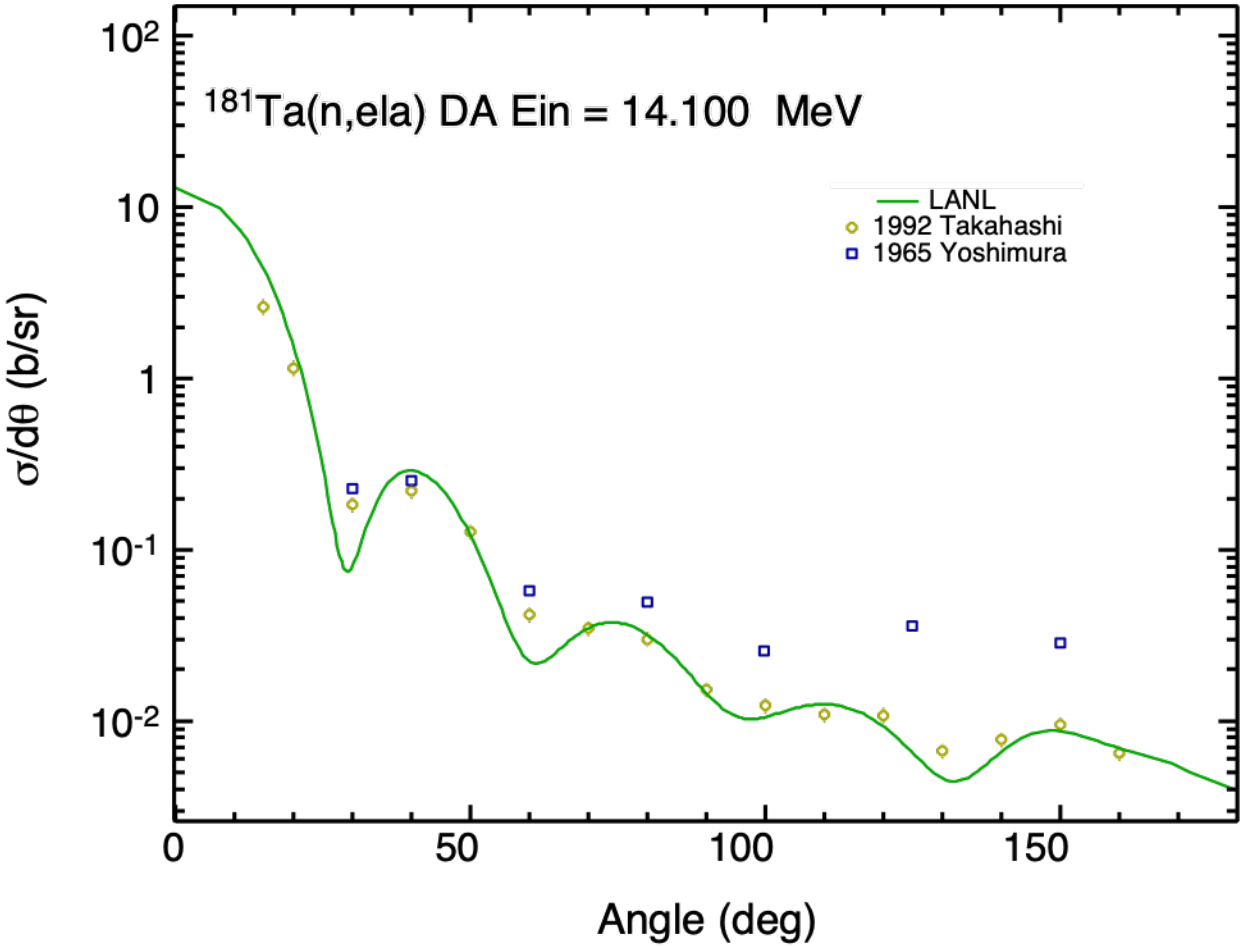

^{181}Ta(n,ela) DA Ein = 6.470 MeV
LANL
1982 Simakov
σ/dθ (b/sr)
10
1
10-1
10-2
0
50
100
150
Angle (deg)

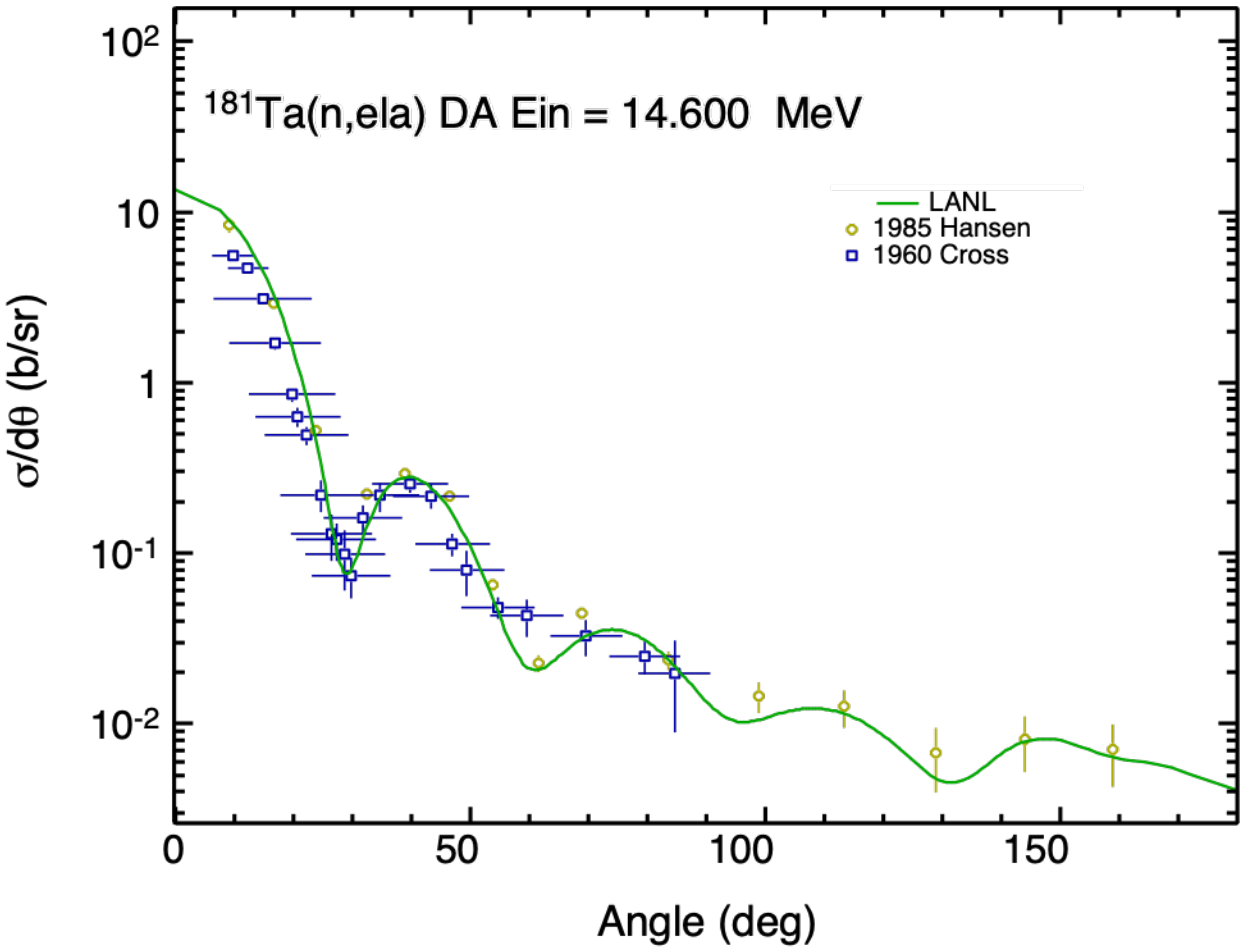

181Ta(n,ela) DA Ein = 7.490 MeV
LANL
1982 Simakov
σ/dθ (b/sr)
10
1
10-1
10-2
0
50
100
150
Angle (deg)

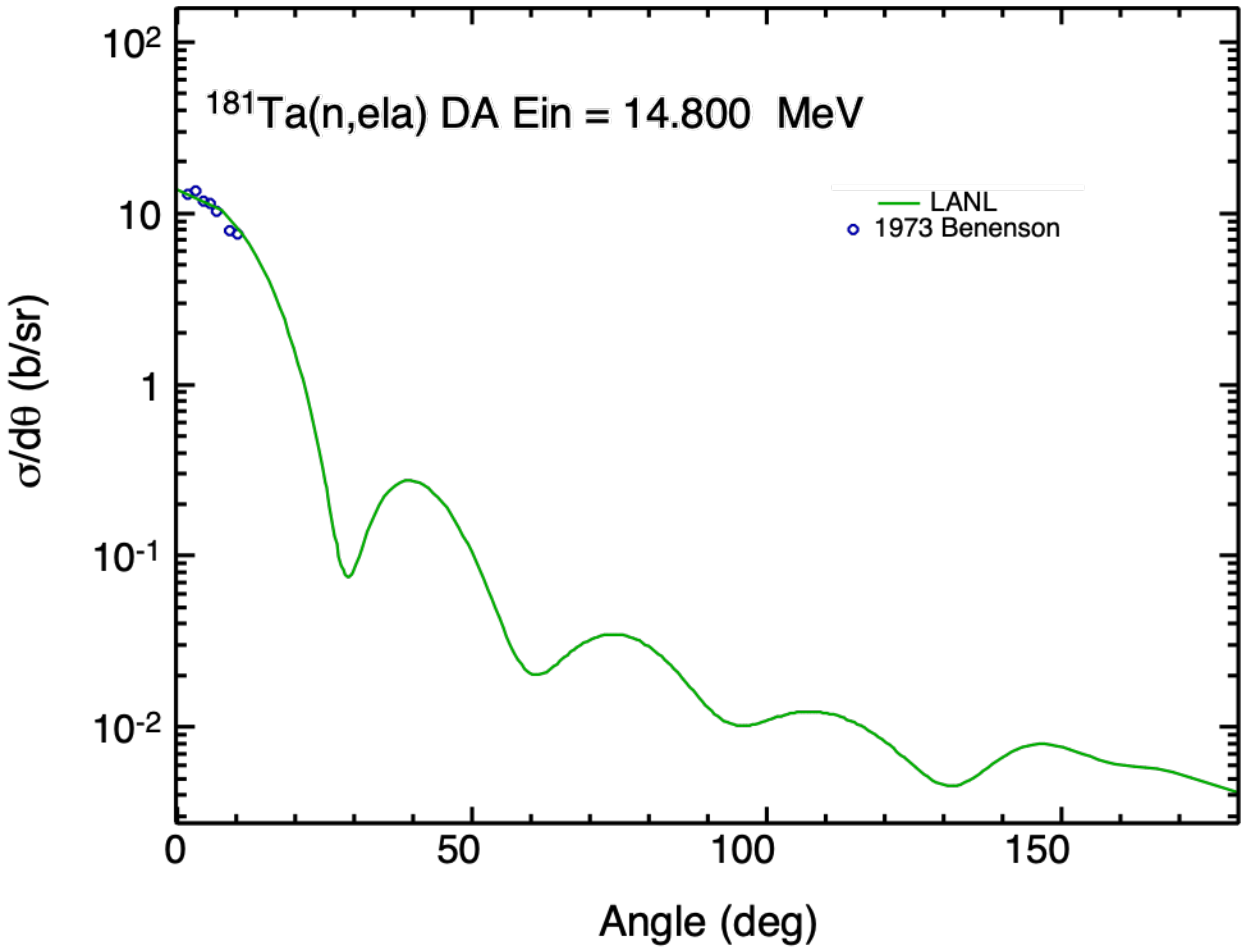

181Ta(n,ela) DA Ein = 7.940 MeV
LANL
1982 Simakov
σ/dθ (b/sr)
20
10
5
2
1
0.5
0.2
10-1
5·10-2
2·10-2
0
50
100
150
Angle (deg)

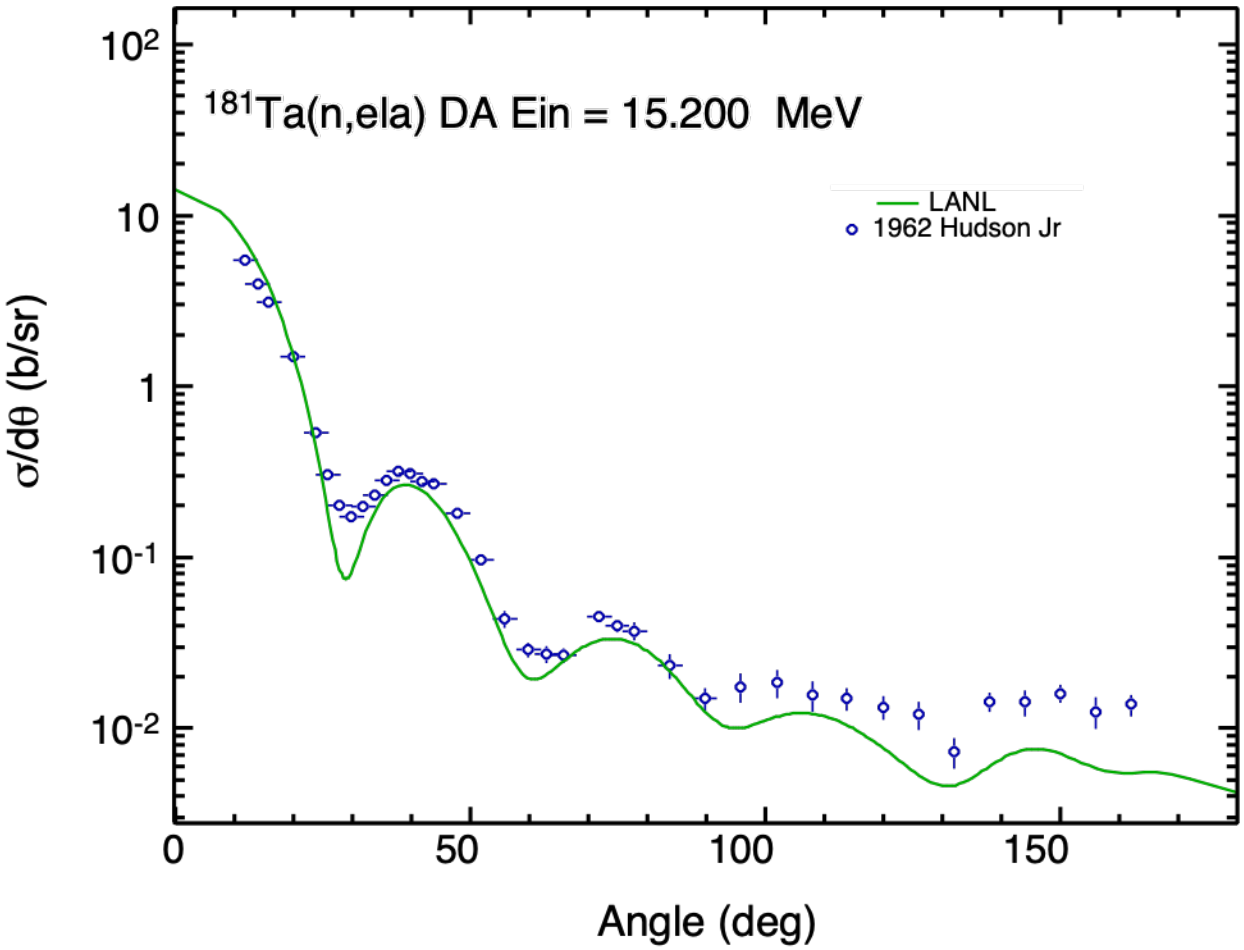

^{181}Ta(n,ela) DA Ein = 11.010 MeV
LANL
1977 Ferrer
σ/dθ (b/sr)
10
1
10-1
10-2
0
50
100
150
Angle (deg)


^{181}Ta(n,ela) DA Ein = 14.000 MeV
LANL
1965 Pearlstein
1957 Rosen
1956 Elliot
σ/dθ (b/sr)
Angle (deg)
10²
10
1
10⁻¹
10⁻²
10⁻³
0
50
100
150

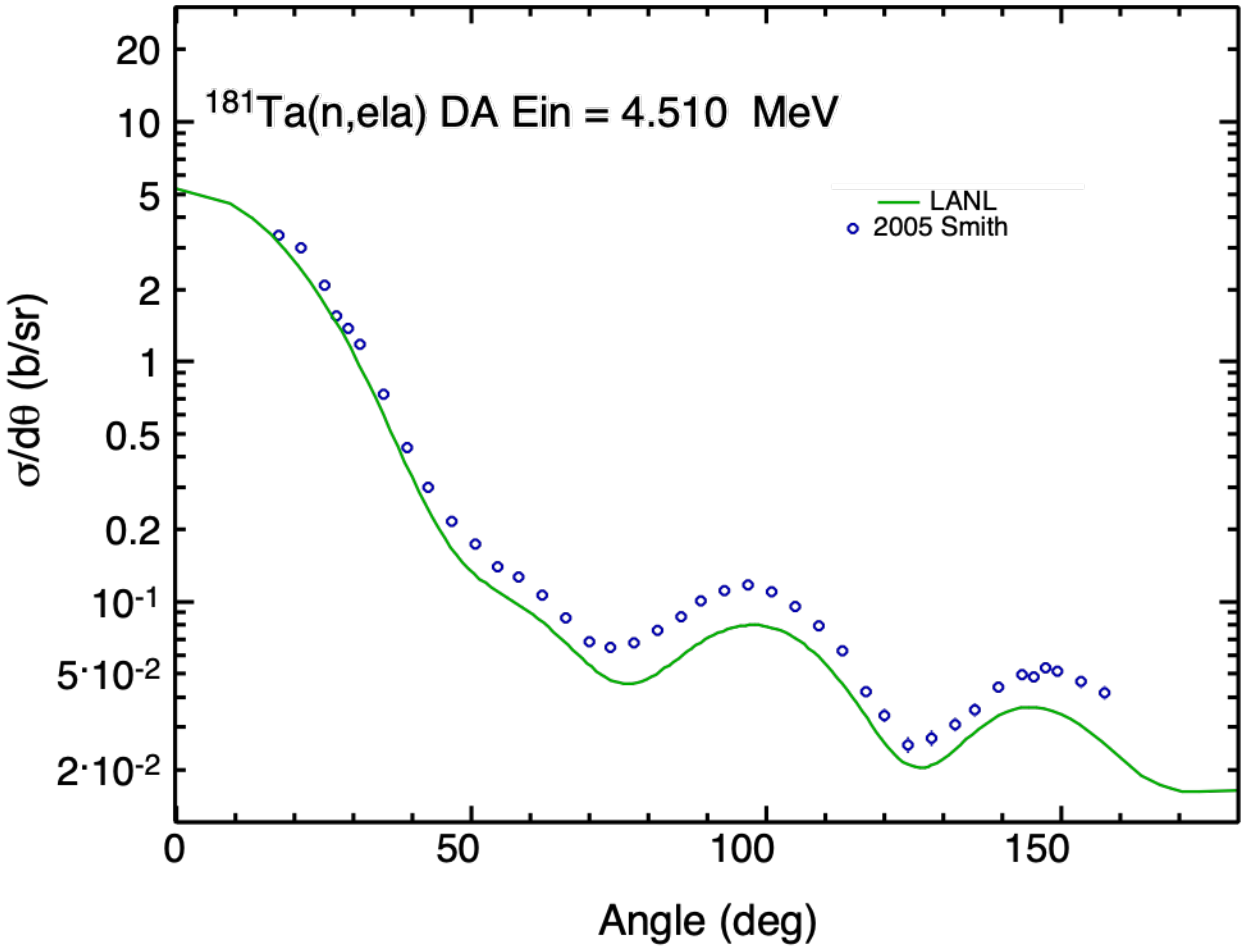

^{181}Ta(n,ela) DA Ein = 14.100 MeV
LANL
1992 Takahashi
1965 Yoshimura
σ/dθ (b/sr)
Angle (deg)
10²
10
1
10⁻¹
10⁻²
0
50
100
150


^{181}Ta(n,ela) DA Ein = 14.600 MeV
LANL
1985 Hansen
1960 Cross
σ/dθ (b/sr)
Angle (deg)
10²
10
1
10⁻¹
10⁻²
0
50
100
150

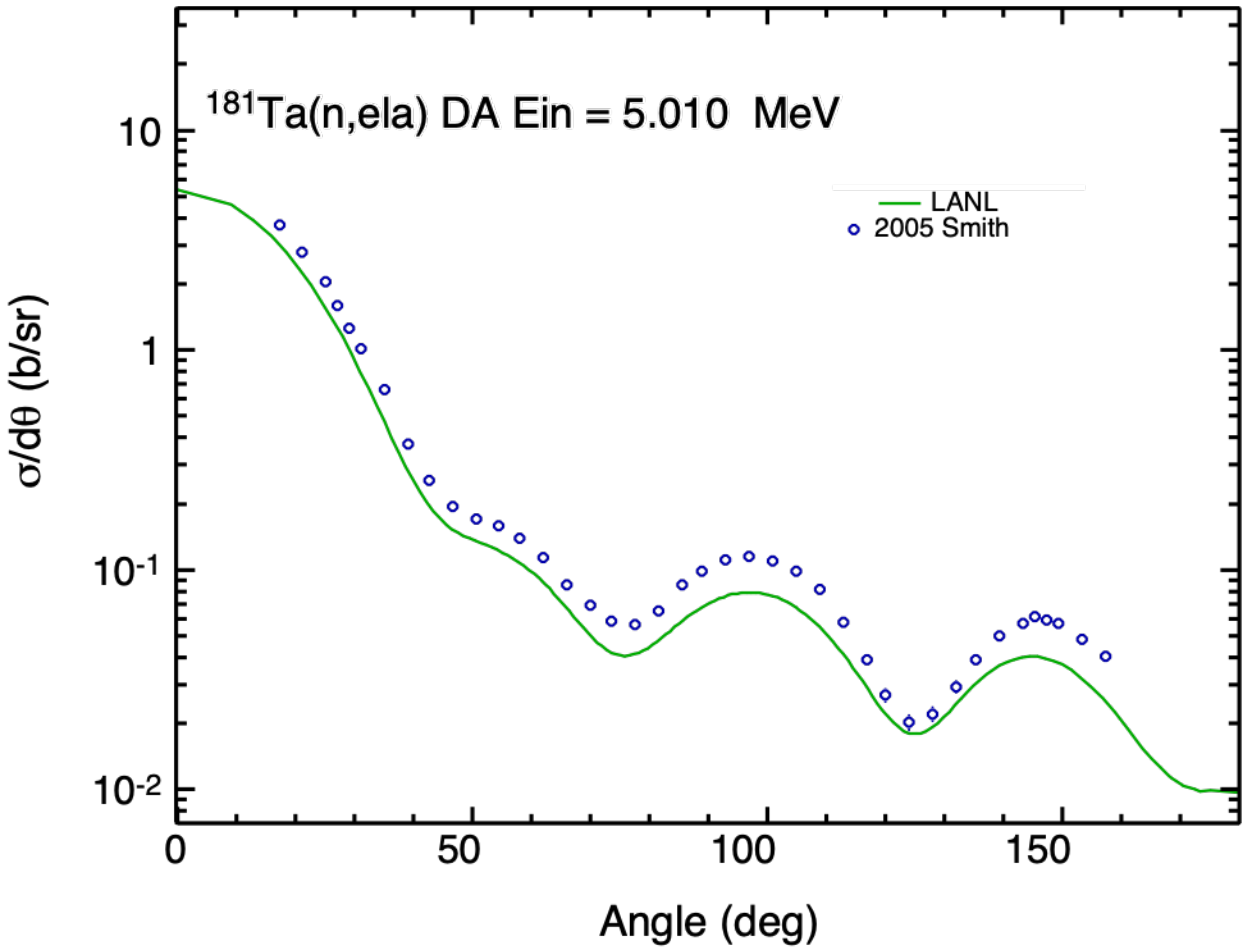

^{181}Ta(n,ela) DA Ein = 14.800 MeV
LANL
1973 Benenson
σ/dθ (b/sr)
Angle (deg)
10²
10
1
10⁻¹
10⁻²
0
50
100
150

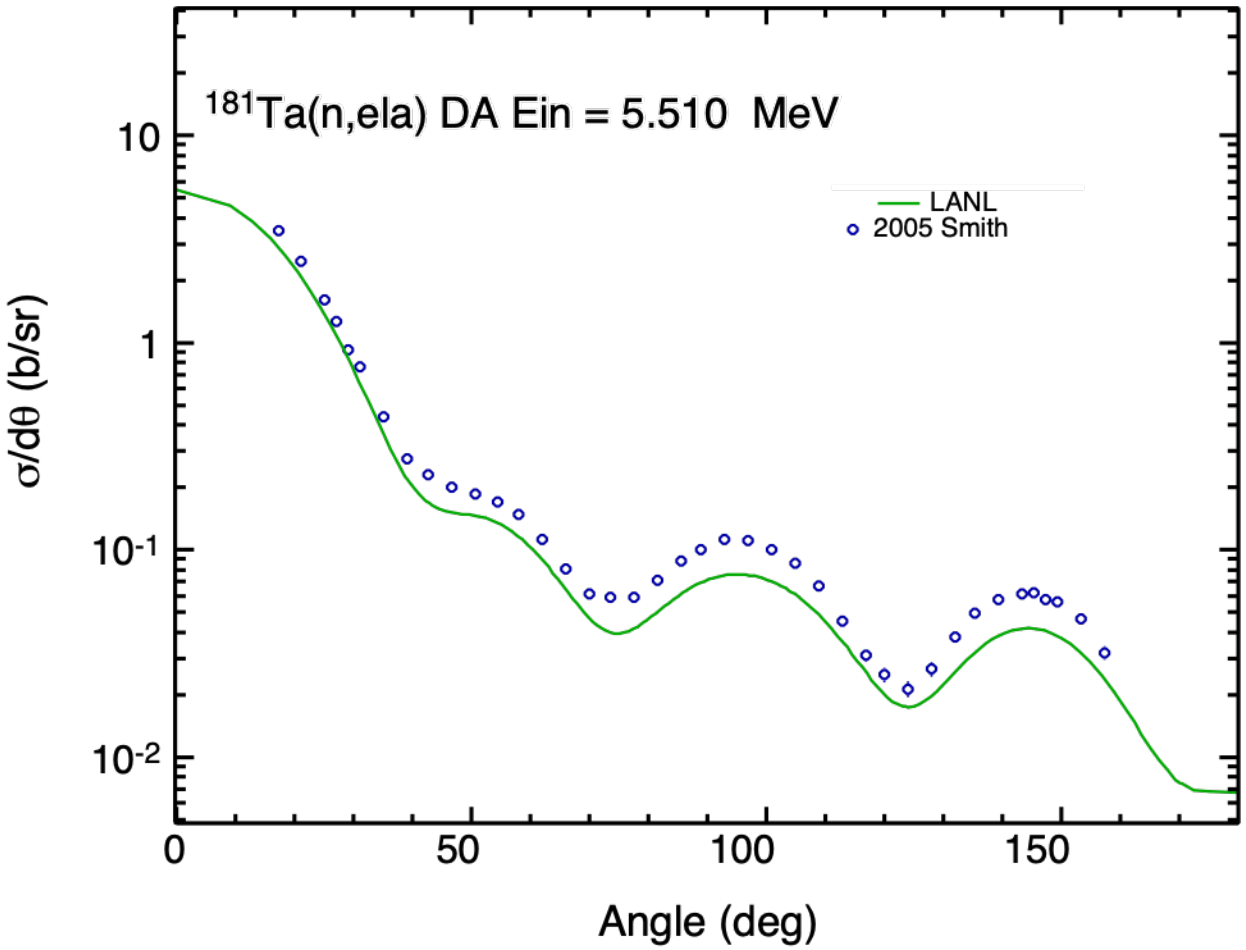

^{181}Ta(n,ela) DA Ein = 15.200 MeV
LANL
1962 Hudson Jr
σ/dθ (b/sr)
Angle (deg)
10²
10
1
10⁻¹
10⁻²
0
50
100
150

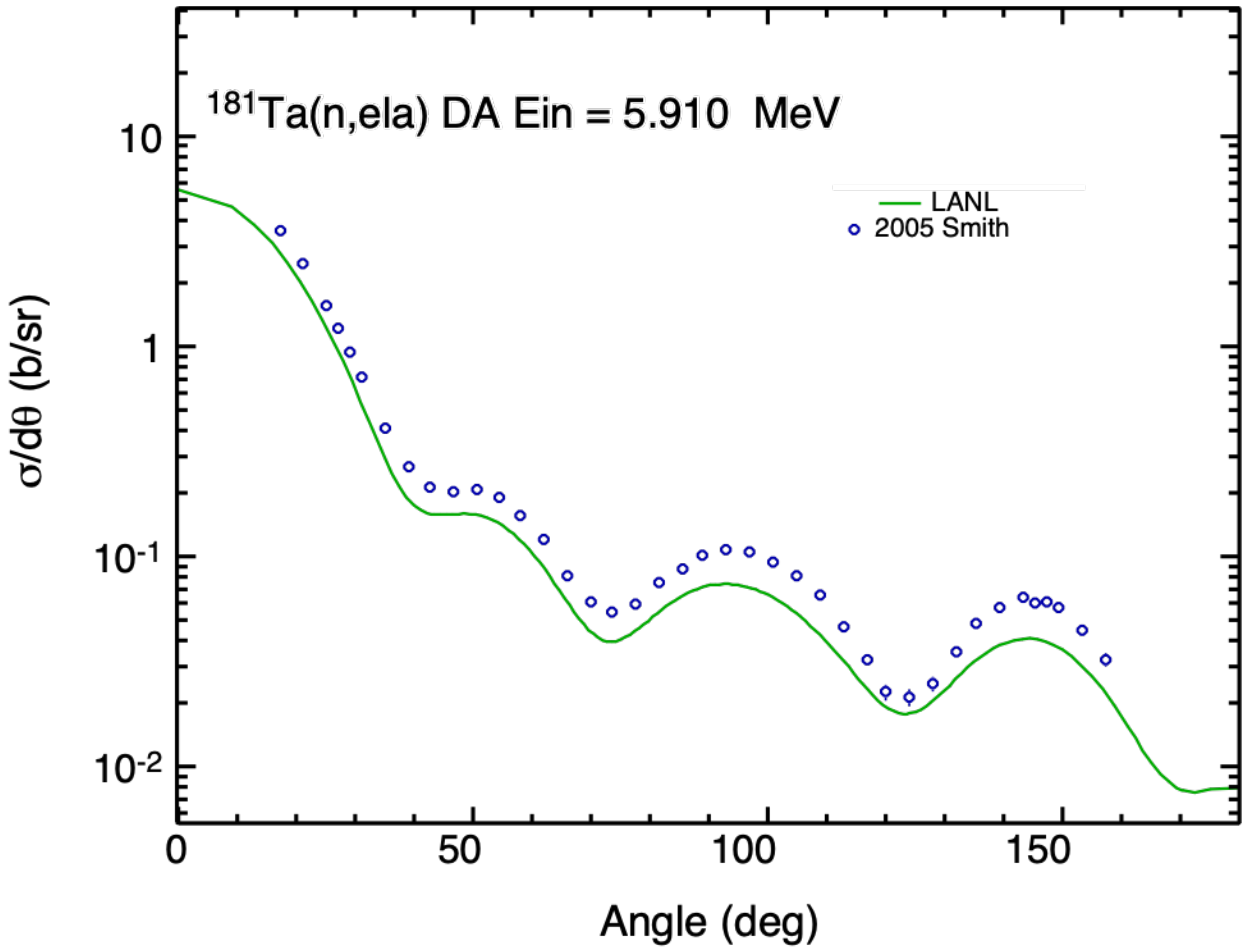
A. Smith: 4.5 - 10 MeV

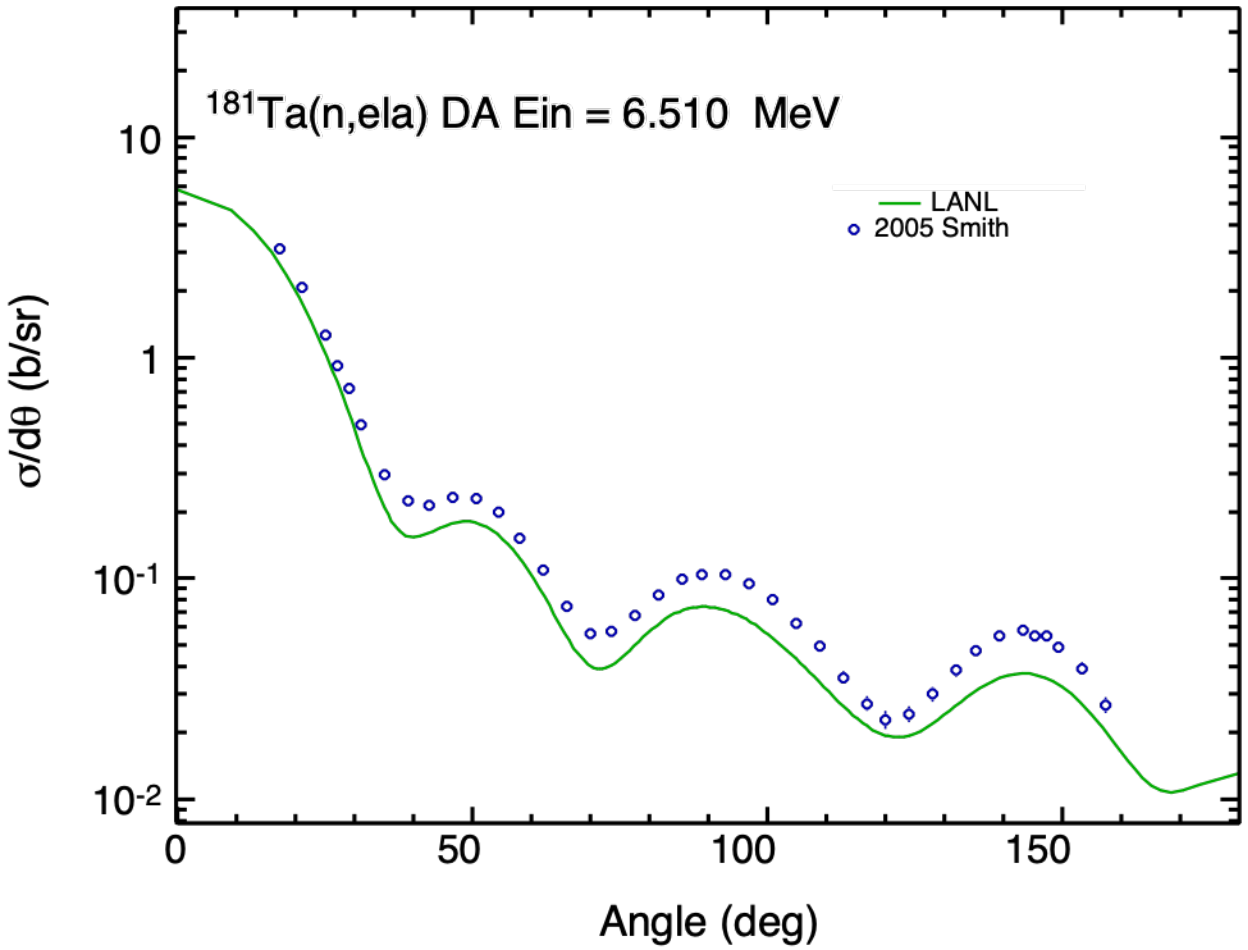


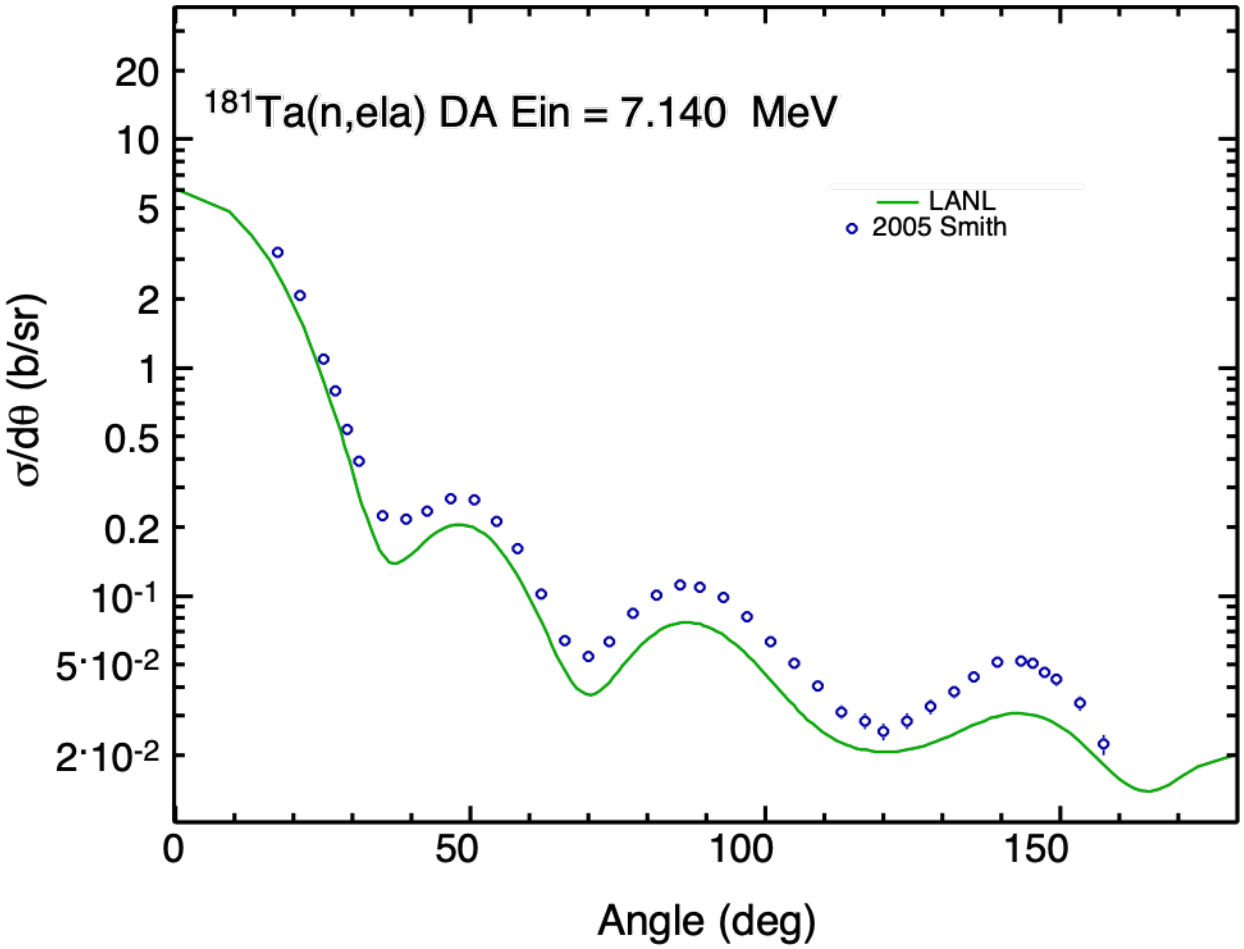
Shape evolution in ~0.5 MeV steps.

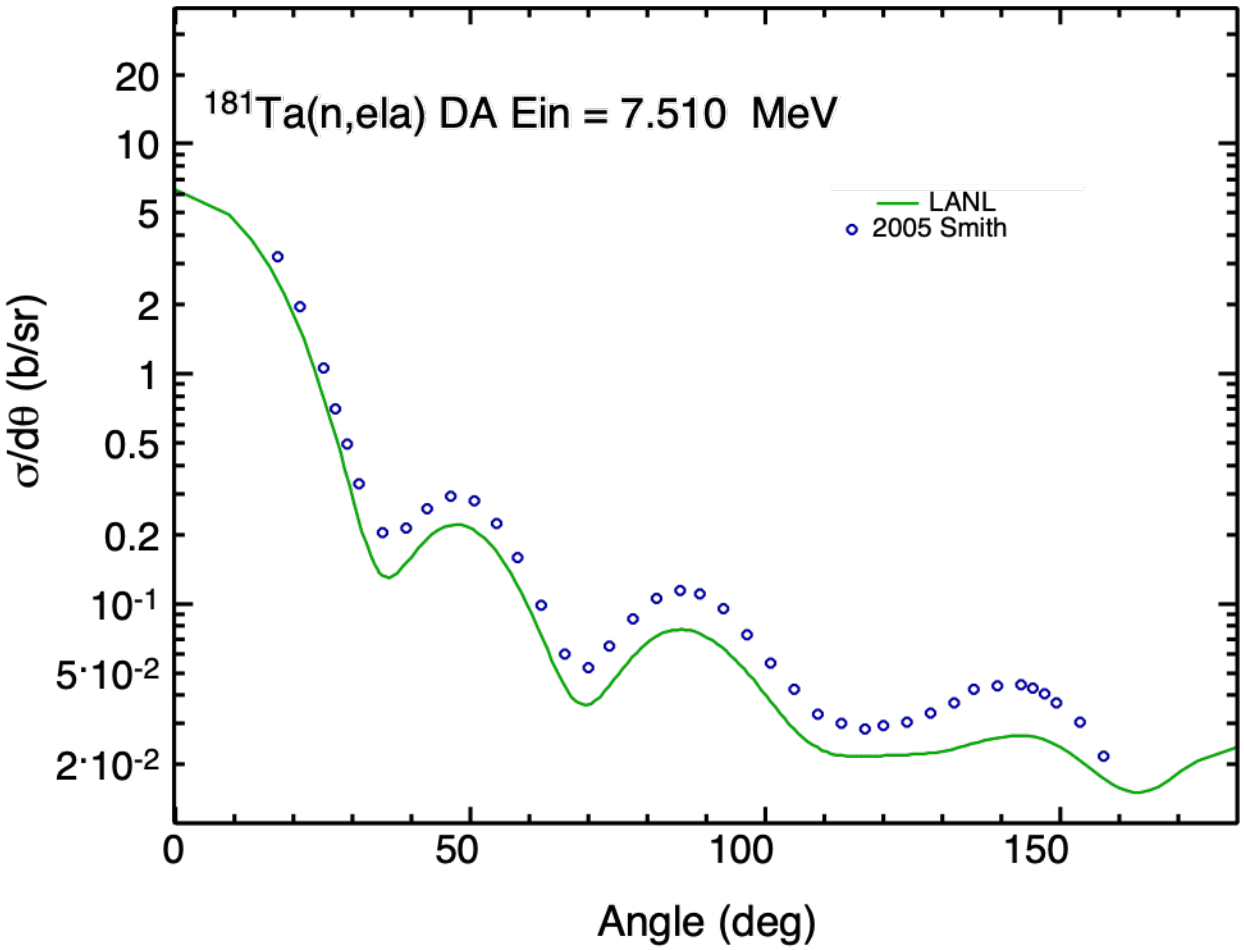

^{181}Ta(n,ela) DA Ein = 5.010 MeV
LANL
2005 Smith
σ/dθ (b/sr)
10
1
10-1
10-2
0
50
100
150
Angle (deg)

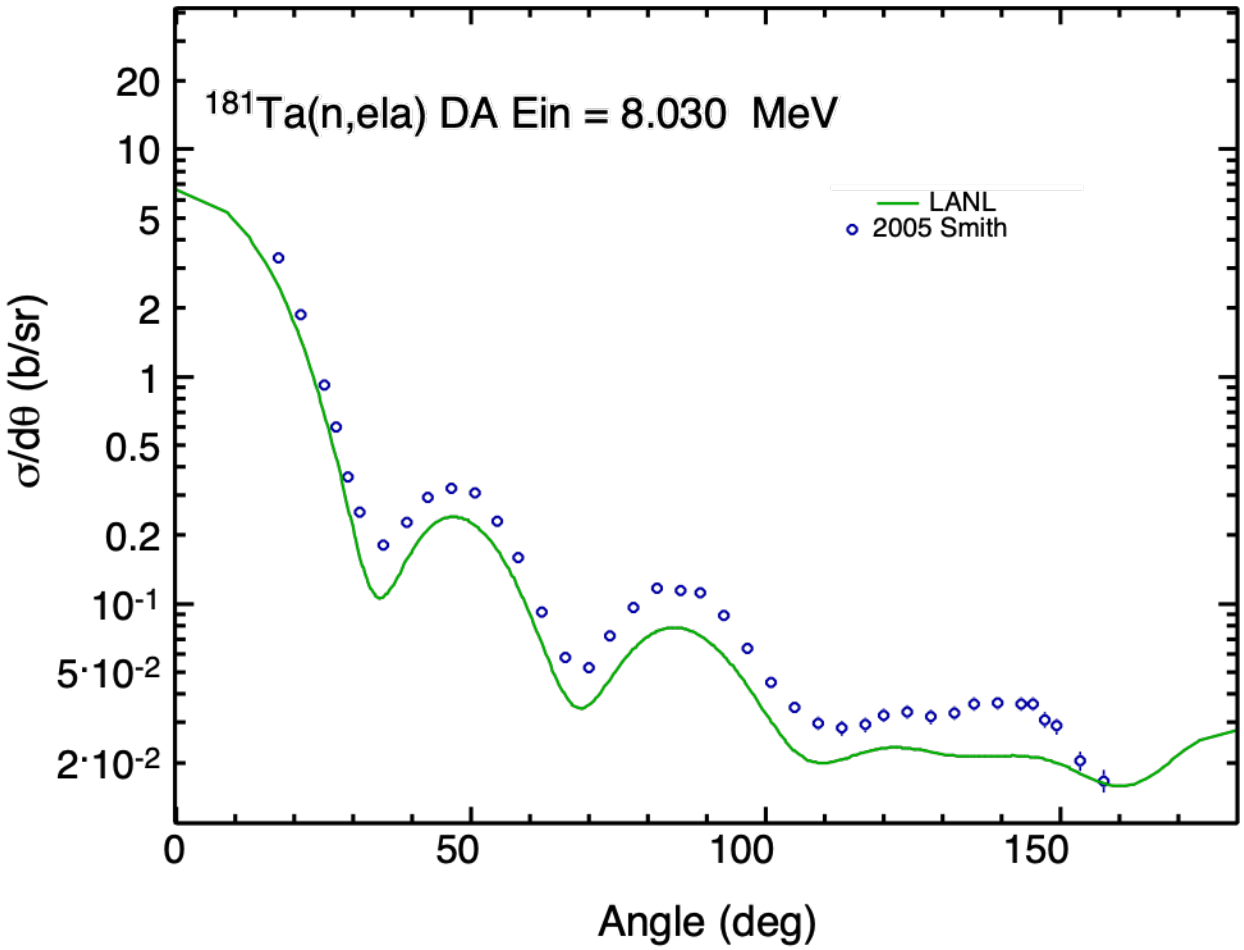

^{181}Ta(n,ela) DA Ein = 5.510 MeV
LANL
2005 Smith
σ/dθ (b/sr)
10
1
10-1
10-2
0
50
100
150
Angle (deg)

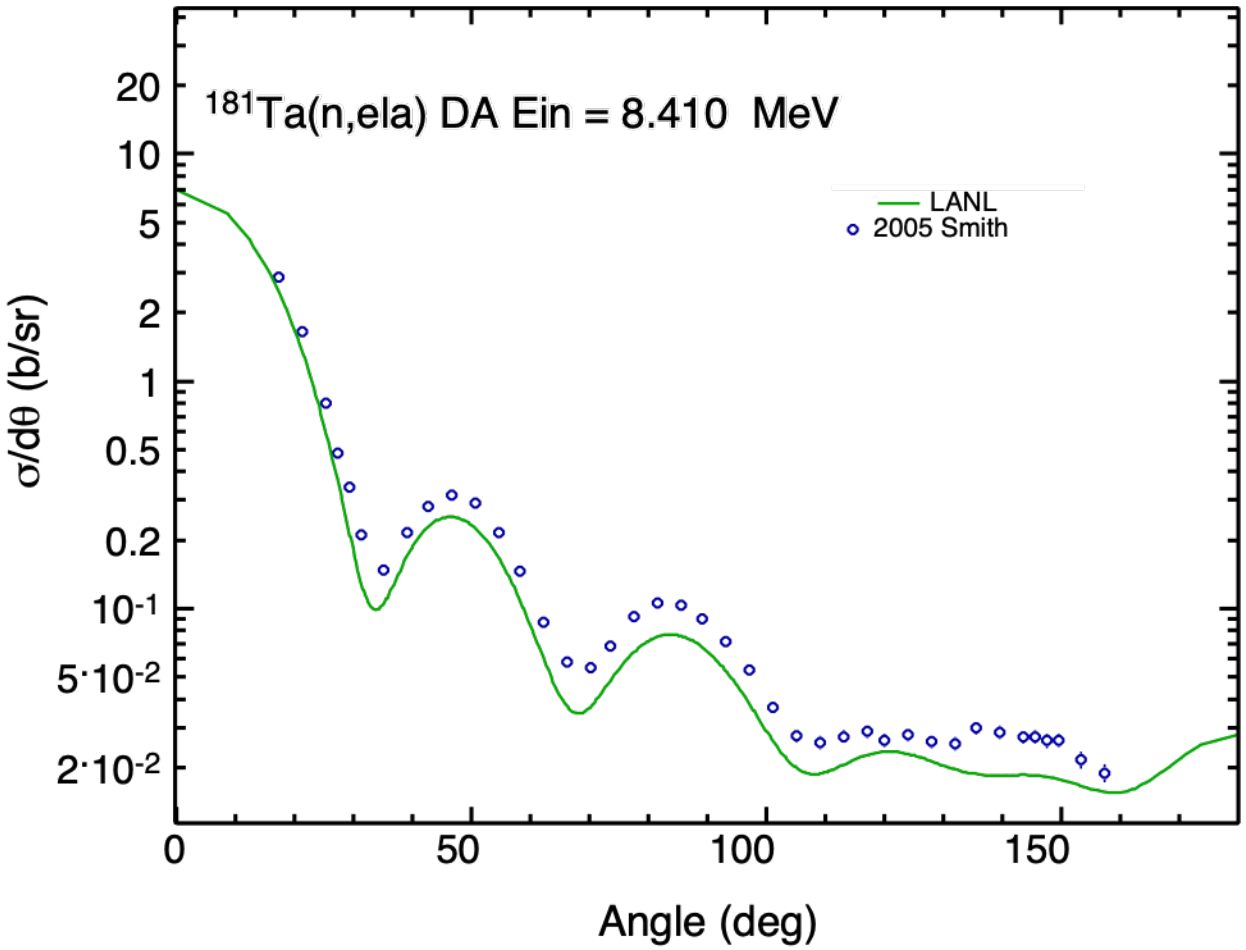

181Ta(n,ela) DA Ein = 5.910 MeV
LANL
2005 Smith
σ/dθ (b/sr)
10
1
10-1
10-2
0
50
100
150
Angle (deg)

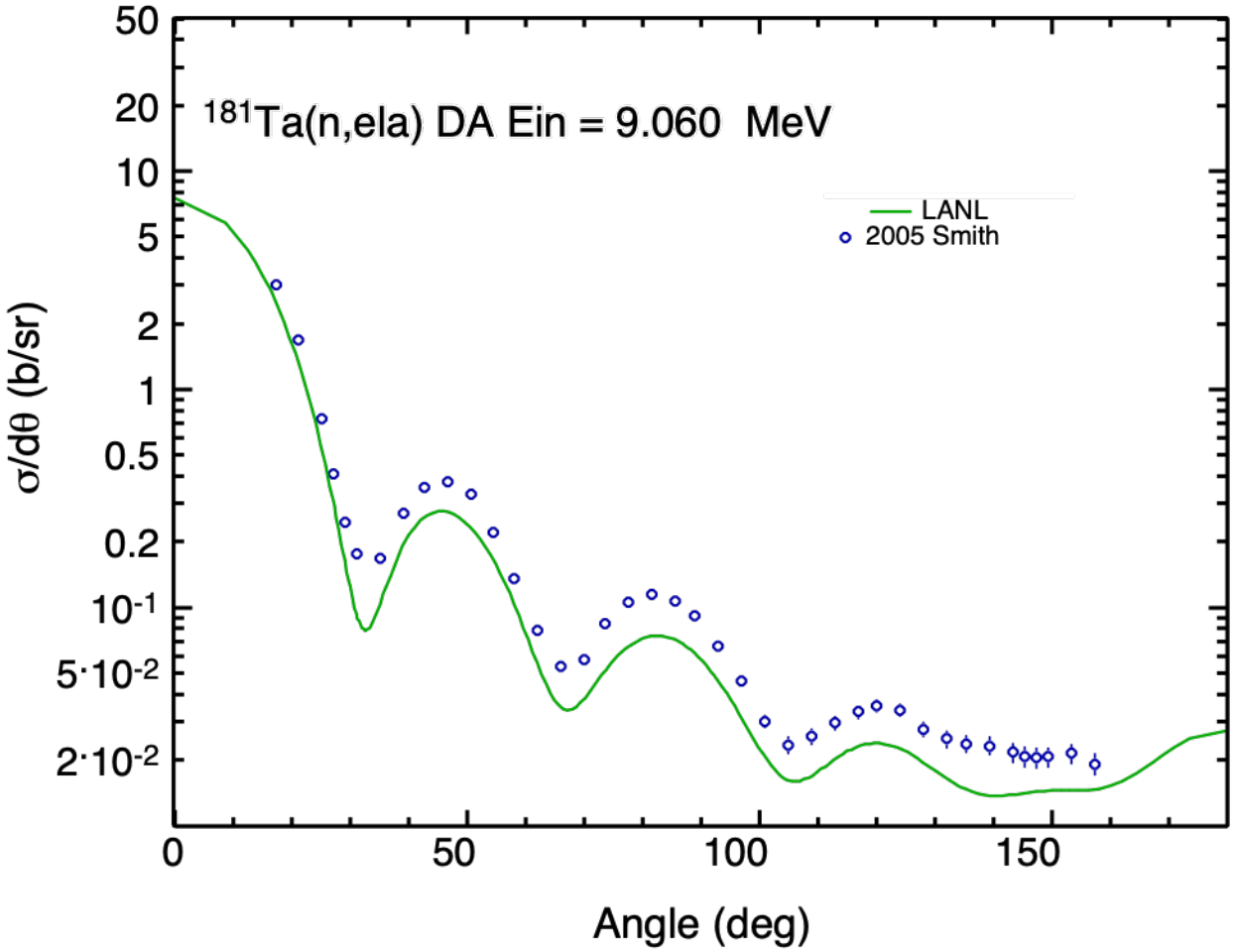

^{181}Ta(n,ela) DA Ein = 6.510 MeV
LANL
2005 Smith
σ/dθ (b/sr)
10
1
10-1
10-2
0
50
100
150
Angle (deg)

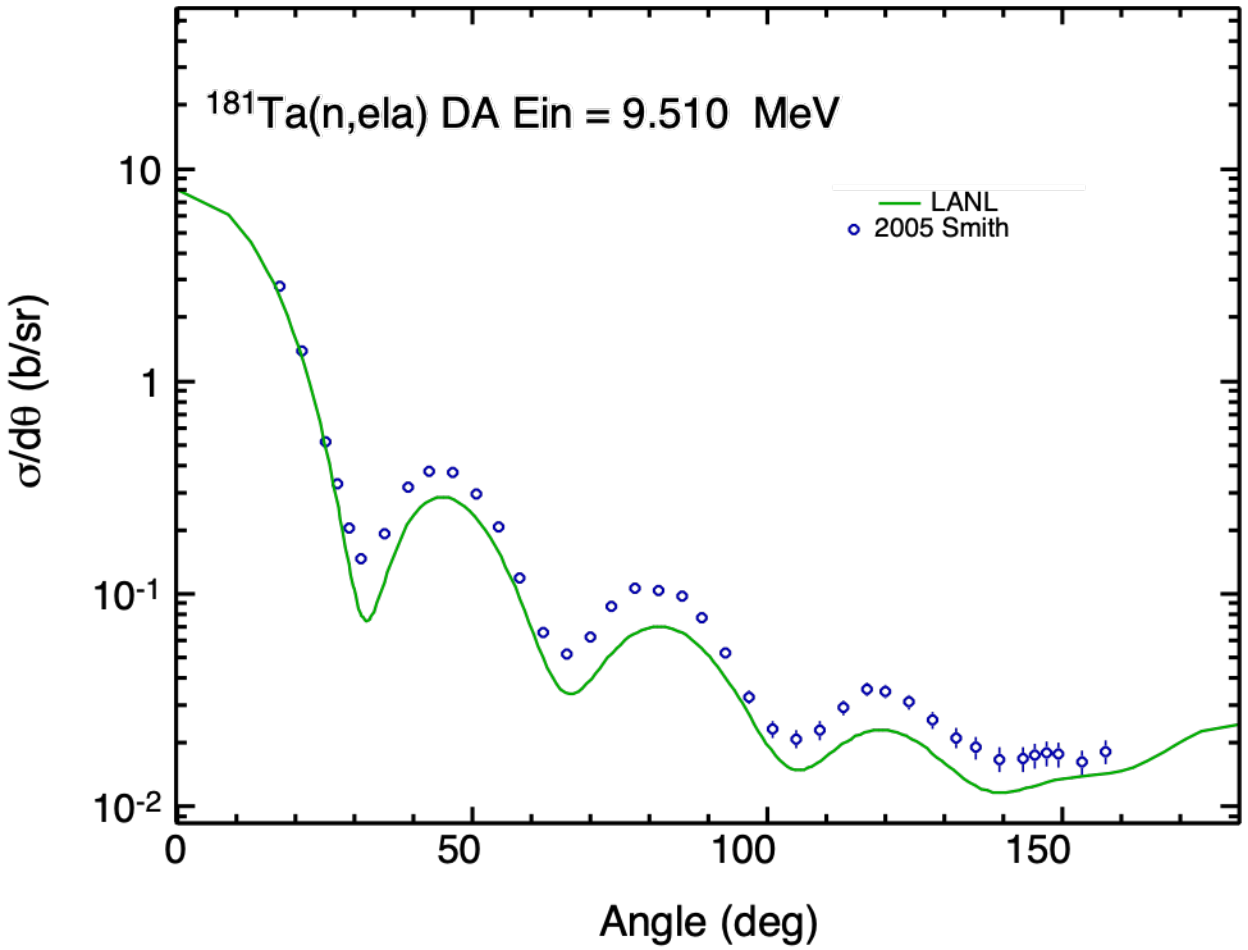

^{181}Ta(n,ela) DA Ein = 7.140 MeV
LANL
2005 Smith
σ/dθ (b/sr)
20
10
5
2
1
0.5
0.2
10-1
5·10-2
2·10-2
0
50
100
150
Angle (deg)

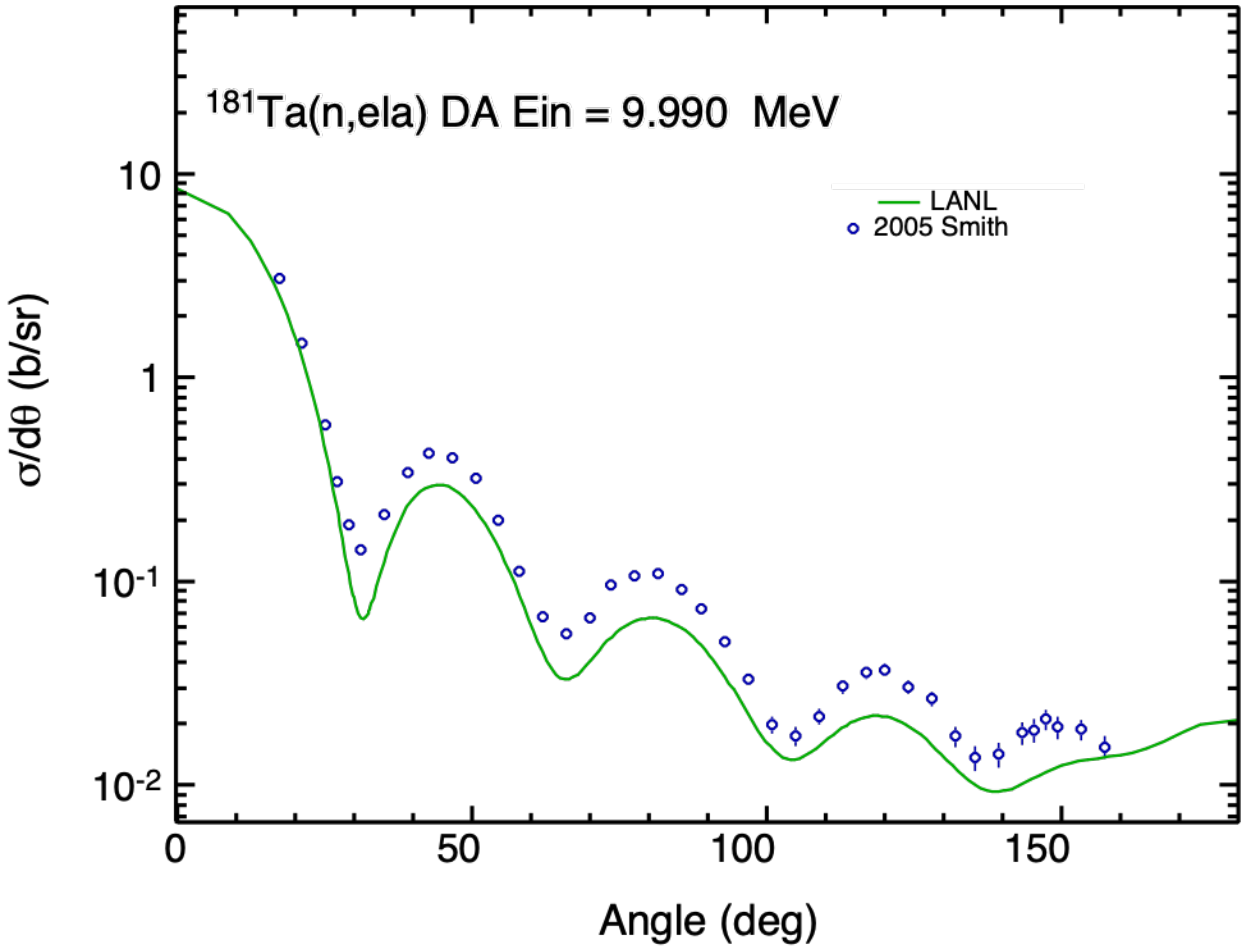

181Ta(n,ela) DA Ein = 7.510 MeV
LANL
2005 Smith
σ/dθ (b/sr)
20
10
5
2
1
0.5
0.2
10-1
5·10-2
2·10-2
0
50
100
150
Angle (deg)


181Ta(n,ela) DA Ein = 8.030 MeV
LANL
2005 Smith
σ/dθ (b/sr)
20
10
5
2
1
0.5
0.2
10-1
5·10-2
2·10-2
0
50
100
150
Angle (deg)

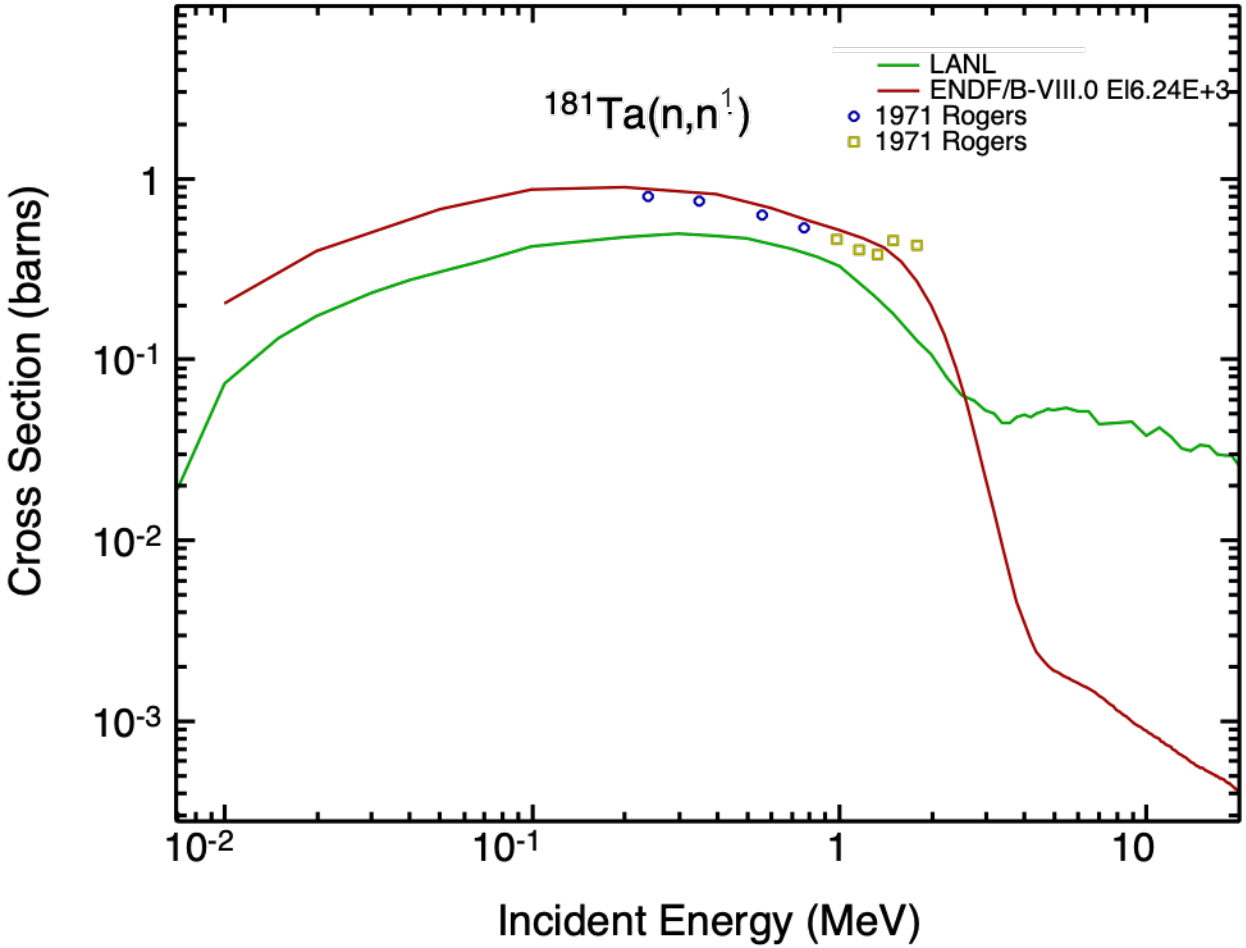

181Ta(n,ela) DA Ein = 8.410 MeV
LANL
2005 Smith
σ/dθ (b/sr)
20
10
5
2
1
0.5
0.2
10-1
5·10-2
2·10-2
0
50
100
150
Angle (deg)


^{181}Ta(n,ela) DA Ein = 9.060 MeV
LANL
2005 Smith
σ/dθ (b/sr)
Angle (deg)
50
20
10
5
2
1
0.5
0.2
10-1
5·10-2
2·10-2
0
50
100
150


^{181}Ta(n,ela) DA Ein = 9.510 MeV
LANL
2005 Smith
σ/dθ (b/sr)
10
1
10-1
10-2
0
50
100
150
Angle (deg)


^{181}Ta(n,ela) DA Ein = 9.990 MeV
LANL
2005 Smith
σ/dθ (b/sr)
10
1
10-1
10-2
0
50
100
150
Angle (deg)

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.

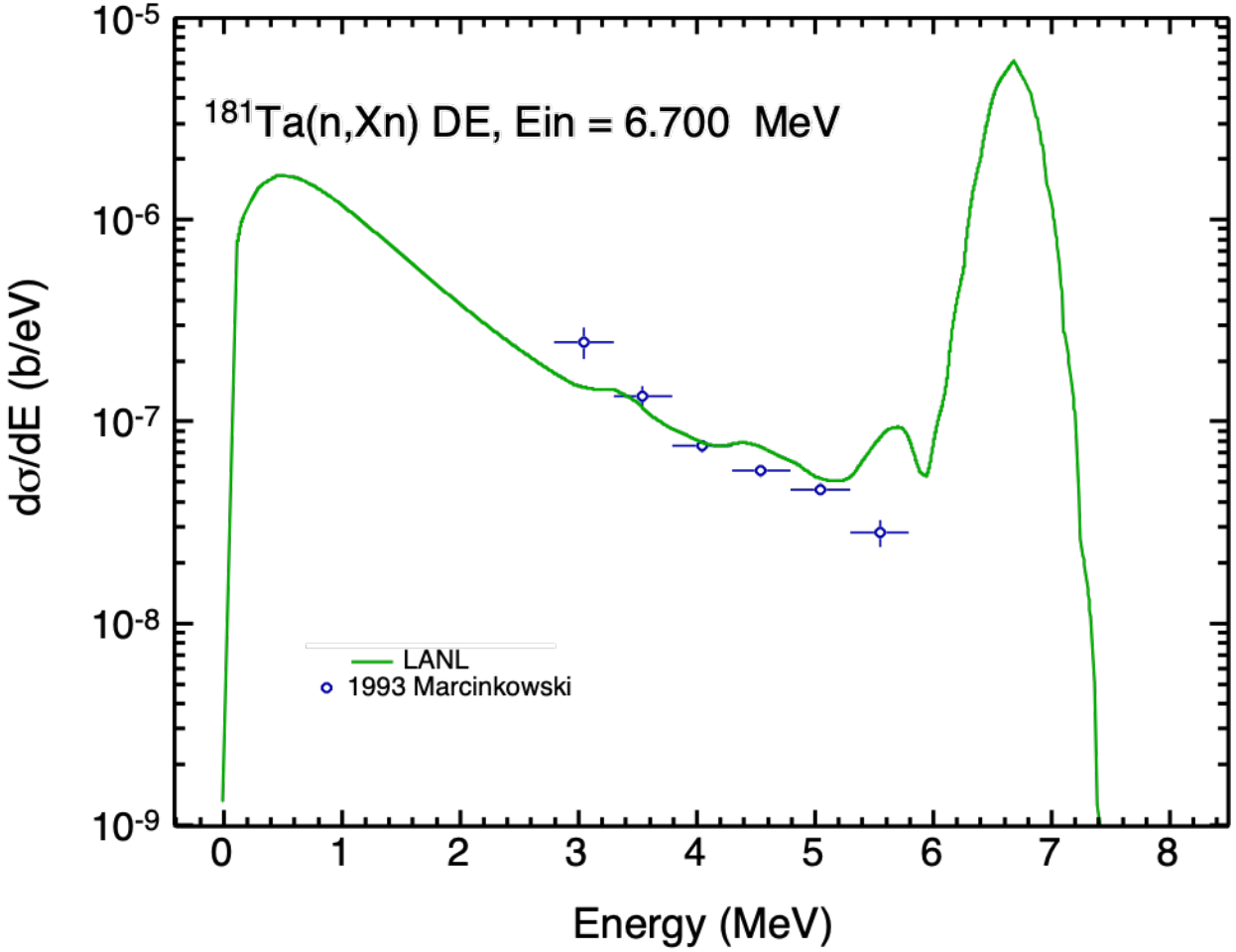
MSD/MSC - neutron spectra, inelastic & (n,xn)

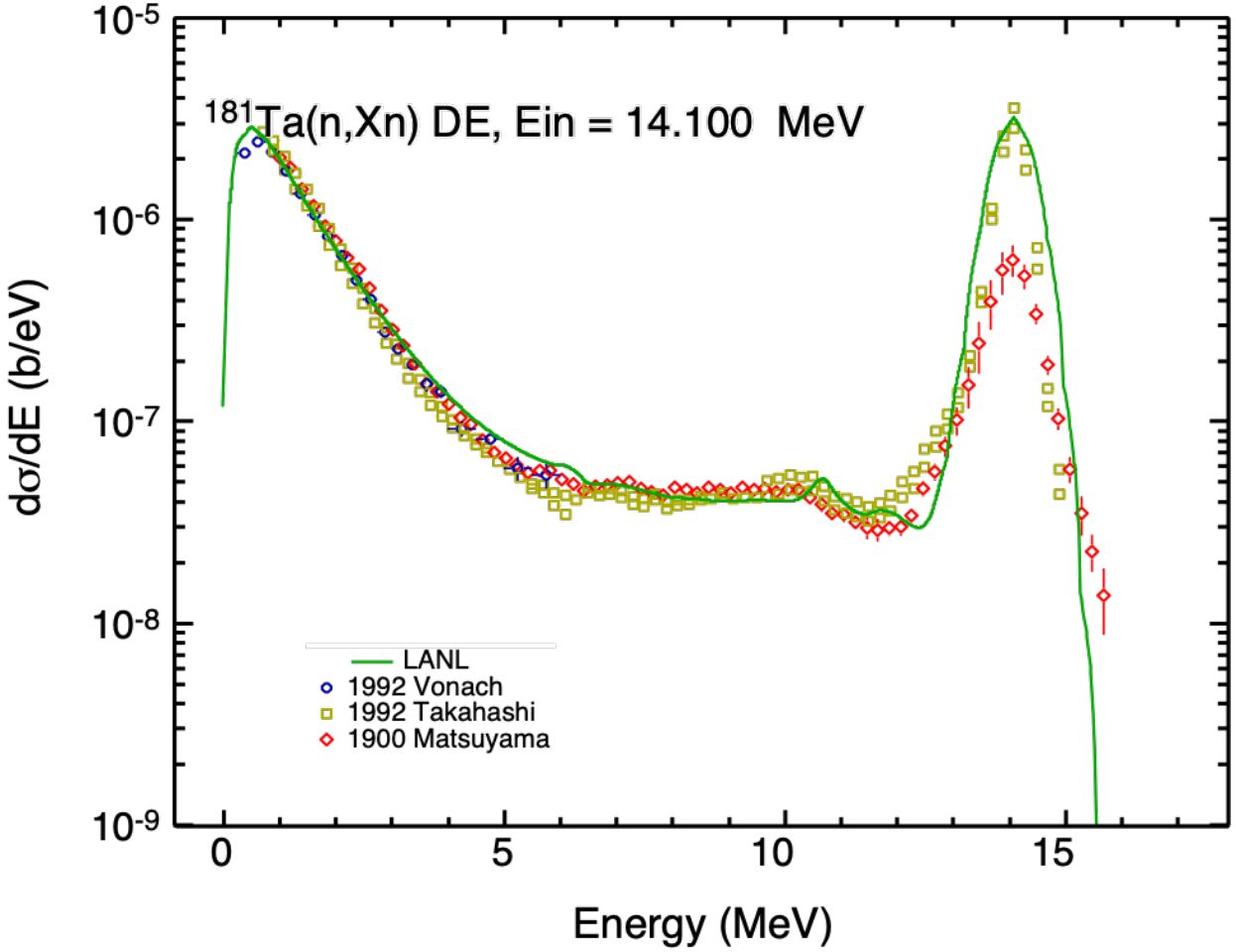
- Quantum mechanical Multistep Direct (MSD) and Multistep Compound (MSC) were employed for neutron emission instead of the exciton model.
- MSD eliminates the need of adding large number of collective discrete levels to the continuum. Default calculations were pretty good but adjusting (manually) a single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

MSD I_o = 6.38 MeV

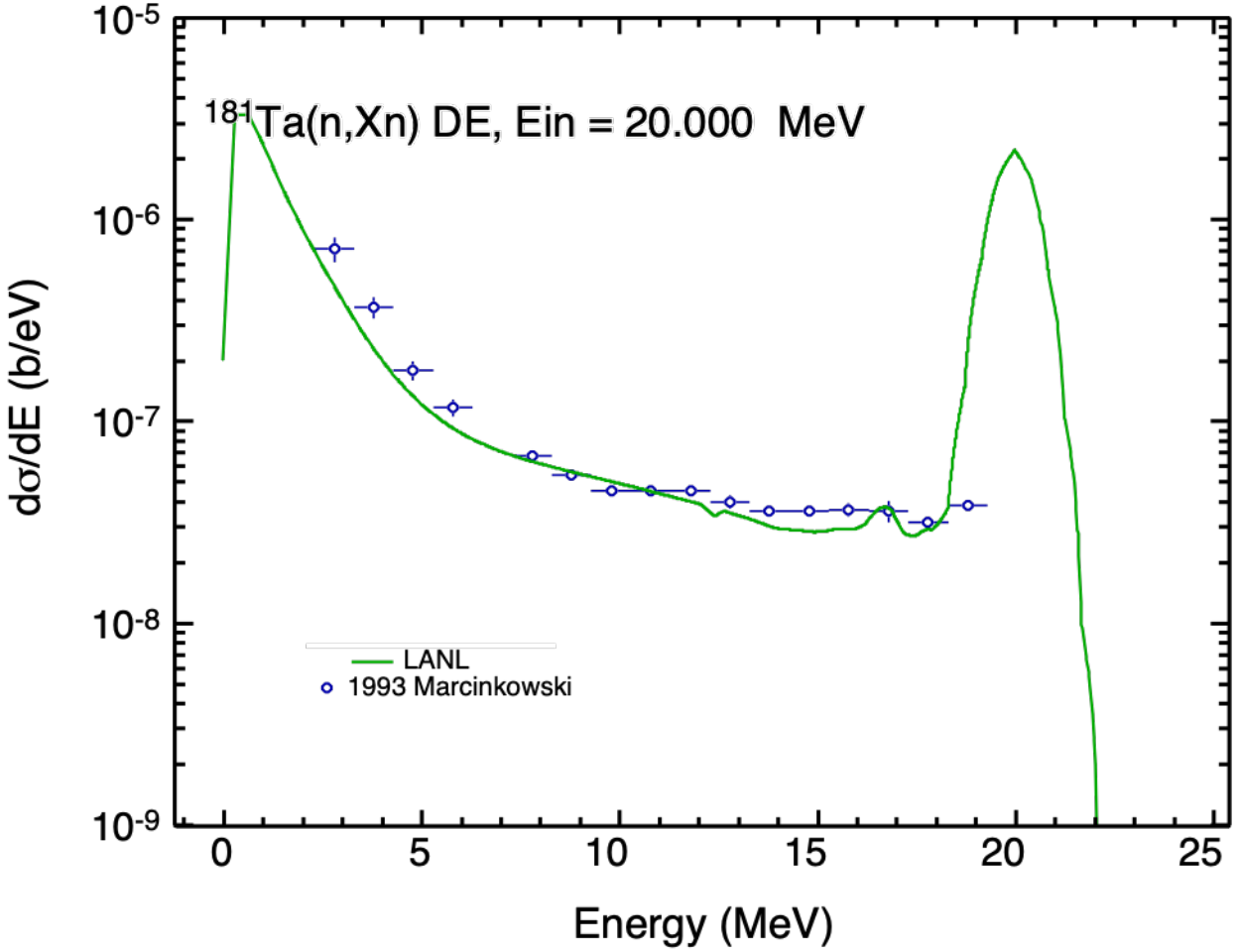
- Extensive experimental coverage of DDX from 5 to 20 MeV of incident energies at different angles was essential for building confidence in the calculations.

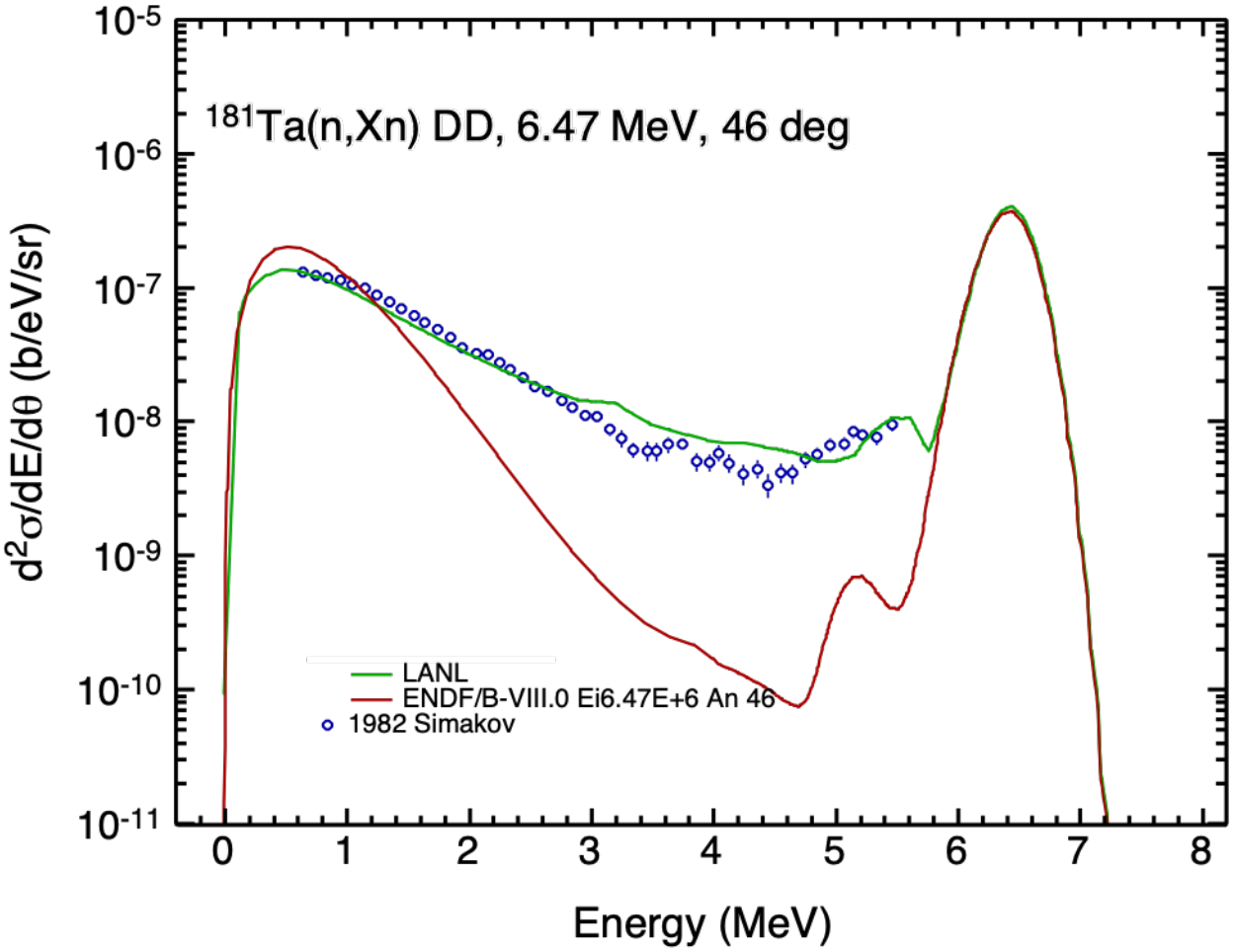
Angle integrated energy n-spectra



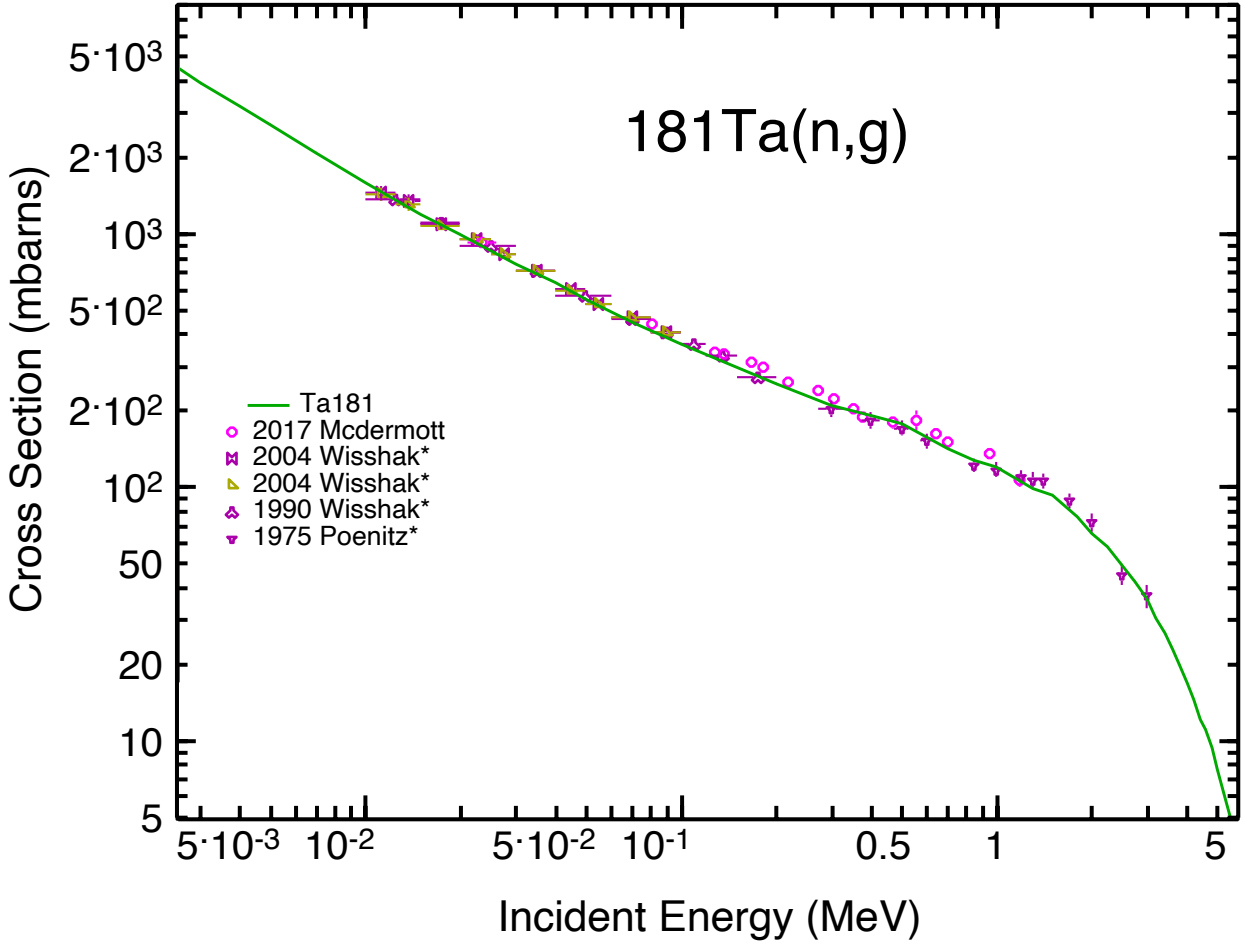


Three consistent data sets are well reproduced by the calculations.


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

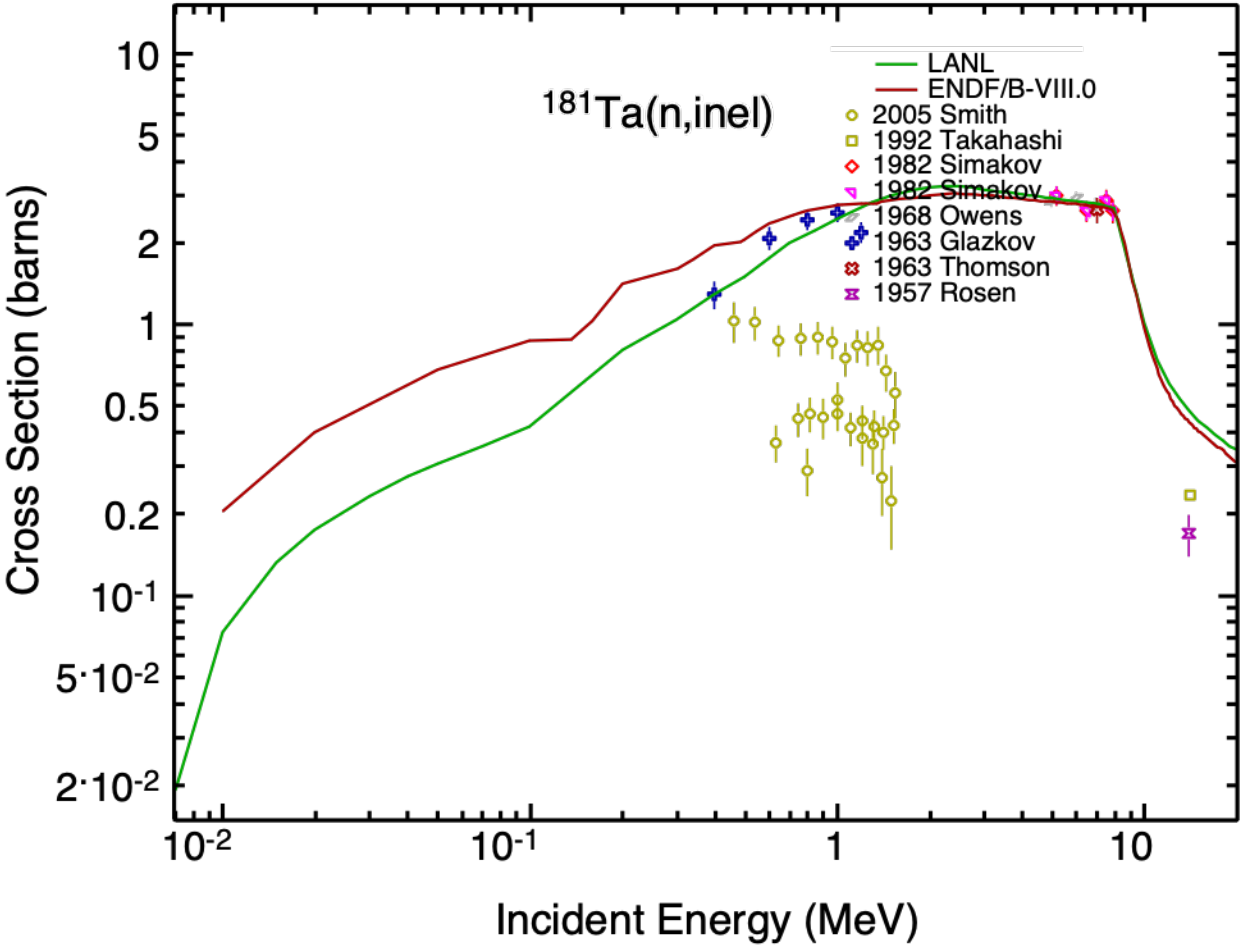

181Ta(n,Xn) DD, 6.47 MeV, 46 deg
d2σ/dE/dθ (b/eV/sr)
Energy (MeV)
LANL
ENDF/B-VIII.0 Ei6.47E+6 An 46
1982 Simakov

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.

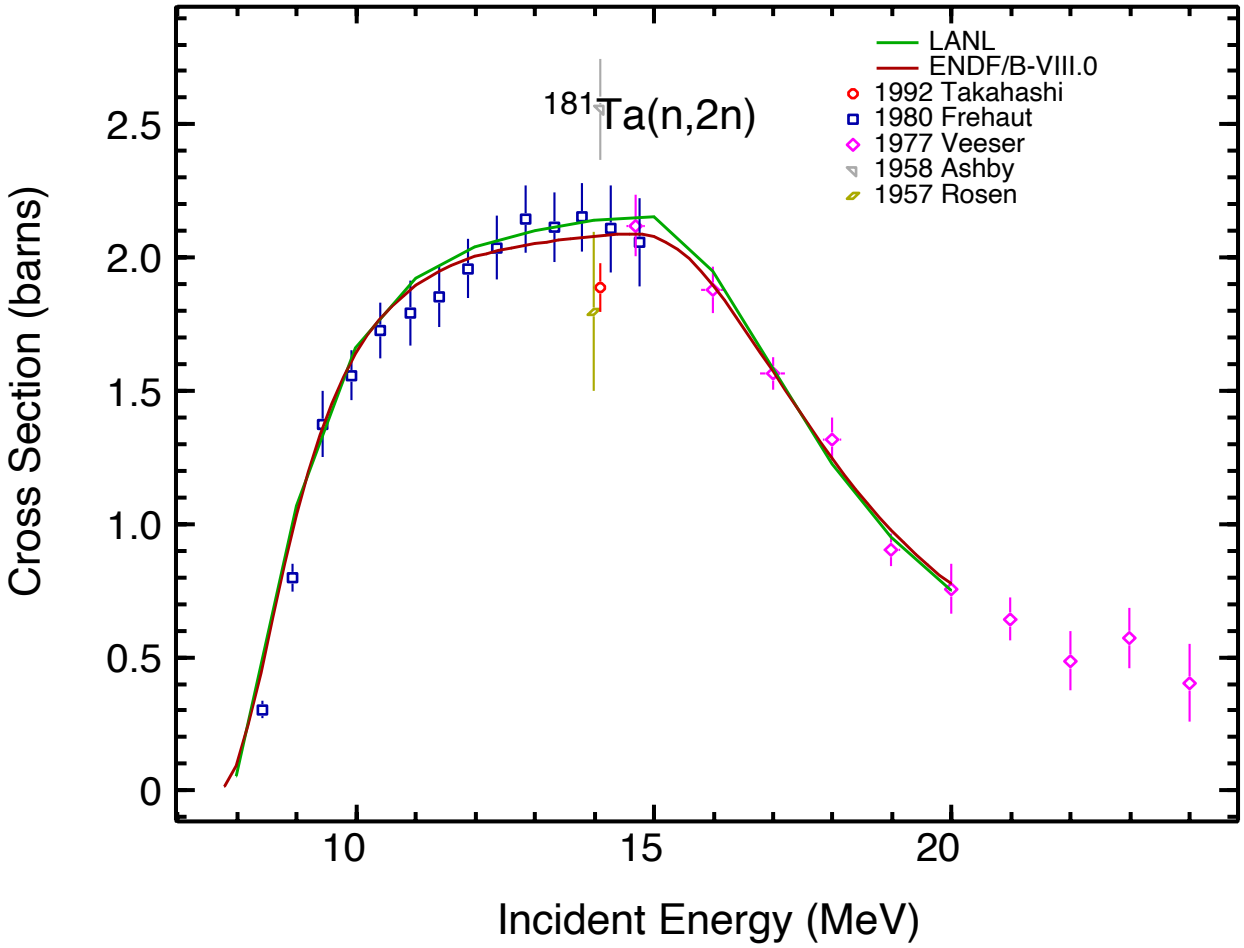
Inelastic cross sections



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

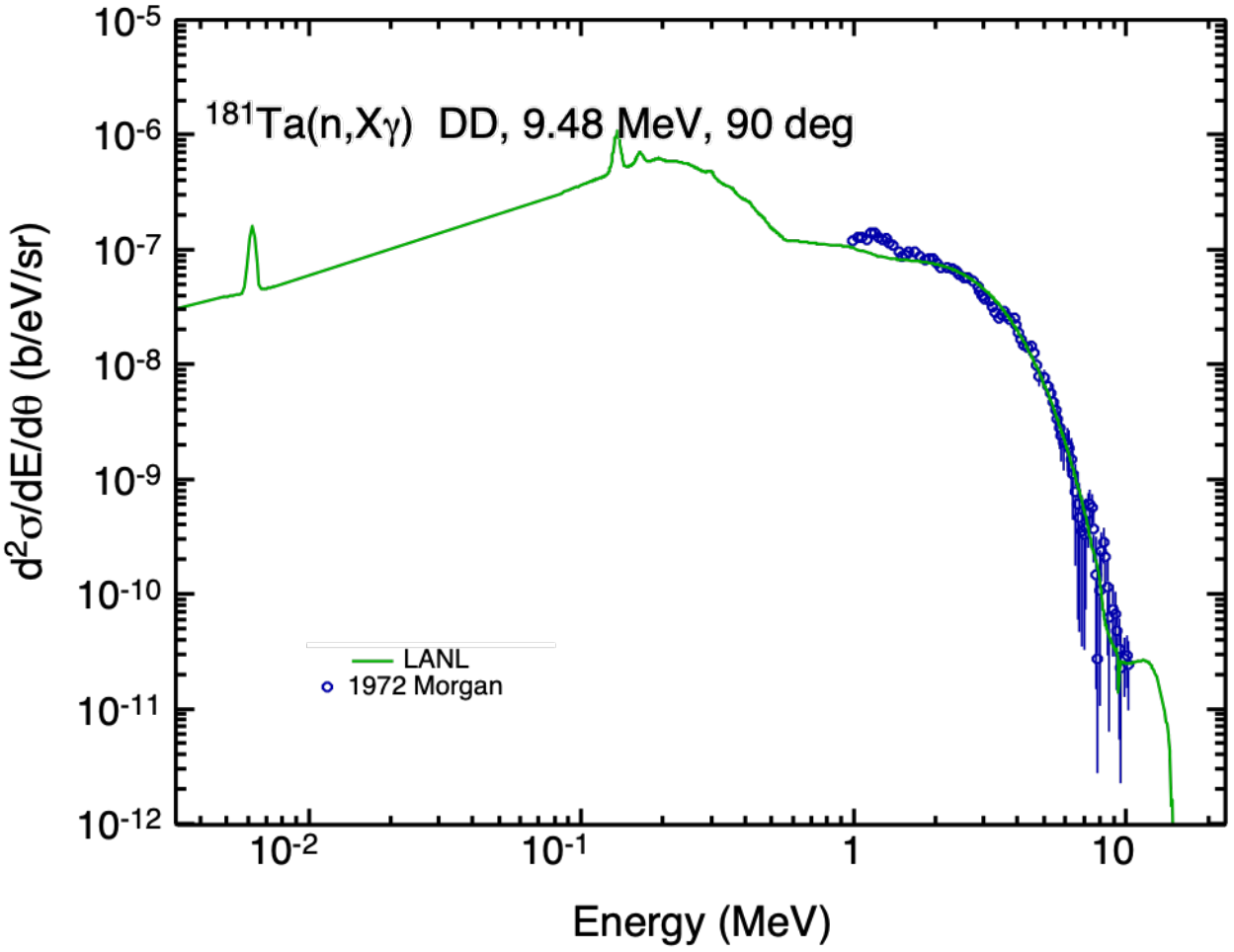
Relatively good agreement with VIII.0.

(n,2n) cross sections



Essentially identical to ENDF/B-VIII.0 but a bit better between 13-14 MeV.

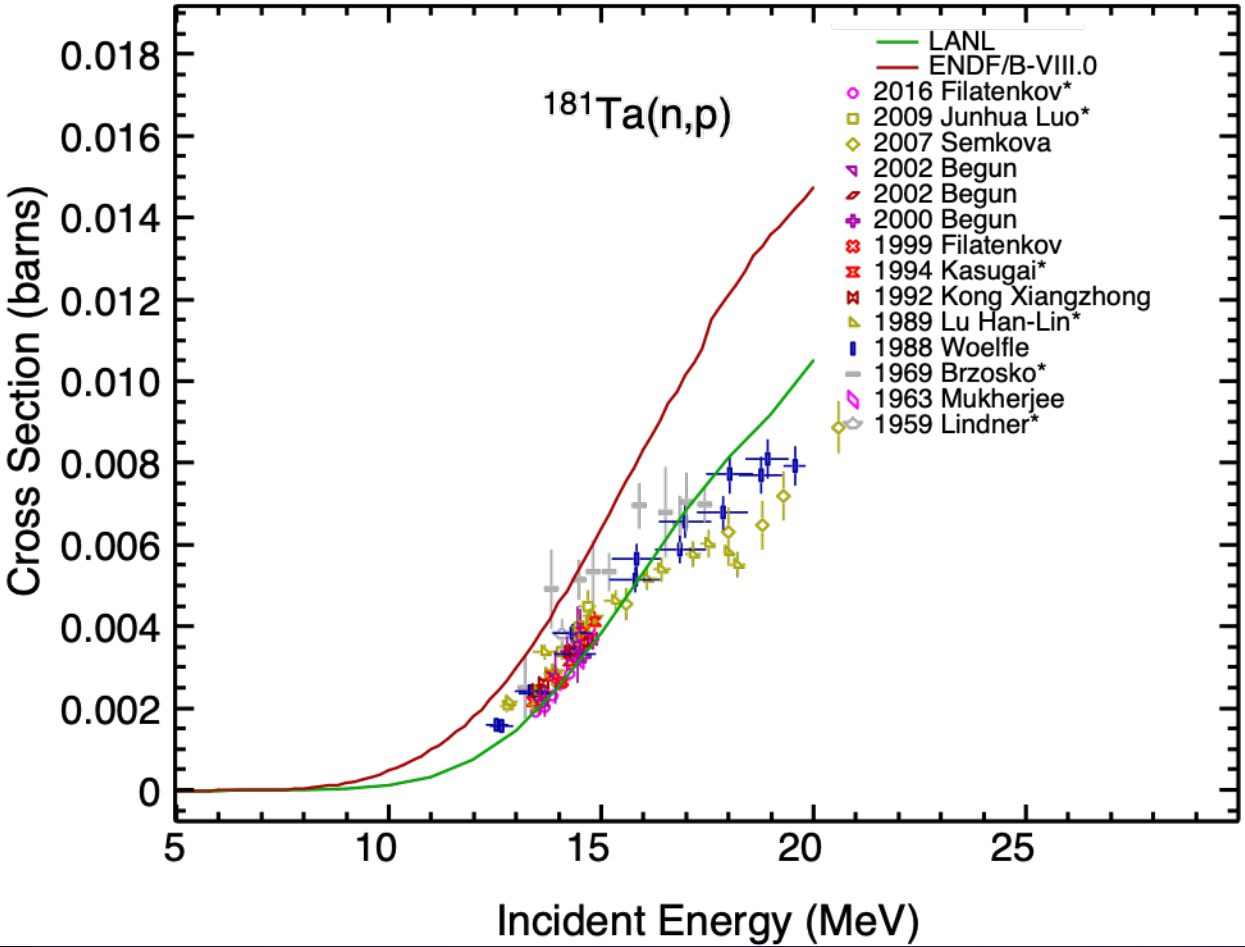
Gamma spectrum



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



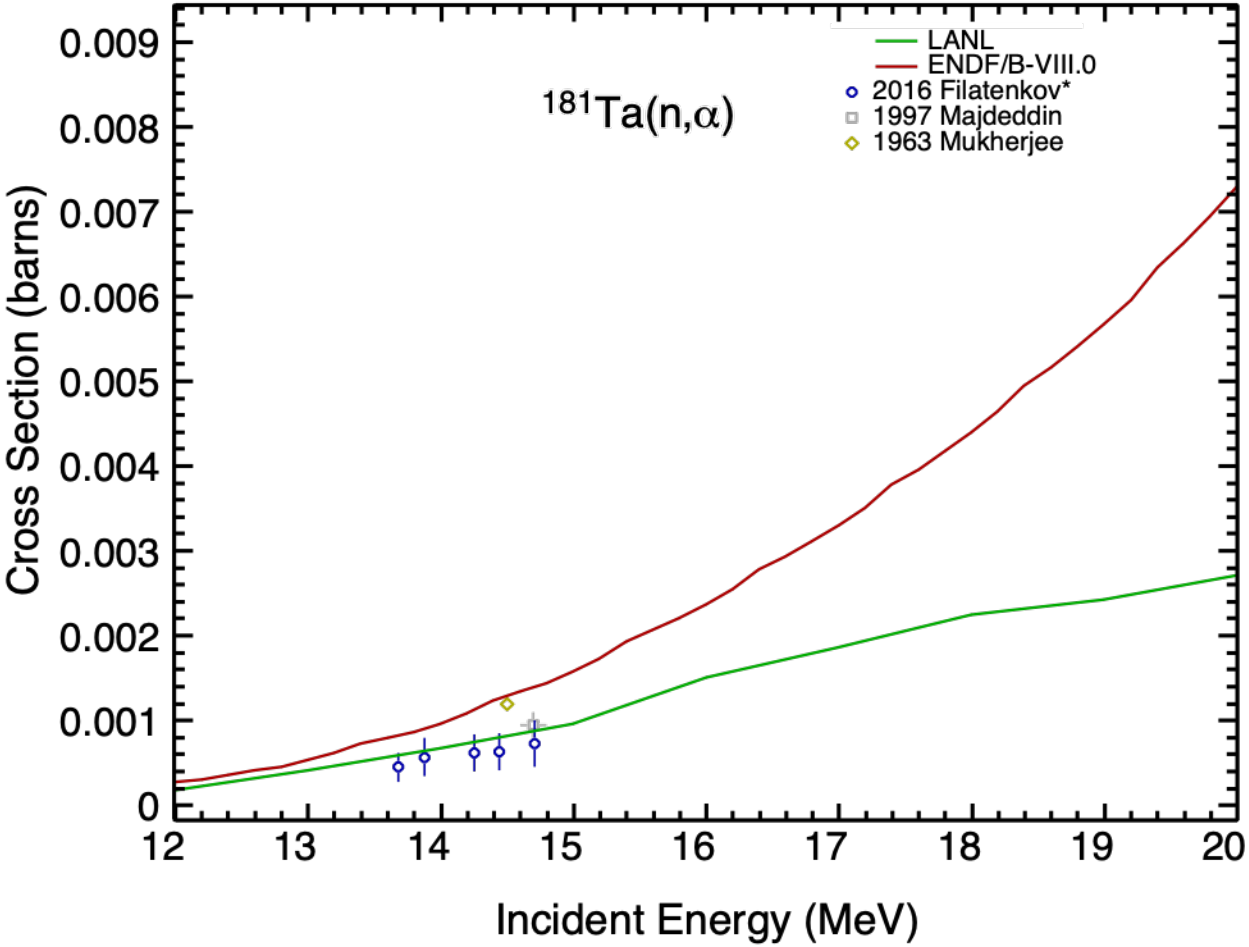
Exciton model is the only option in EMPIRE for charge-exchange reactions and cluster emission in the pre-equilibrium (PE) domain. With neutrons treated within MSD/ MSC there is more freedom to adjust PE yield.

(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

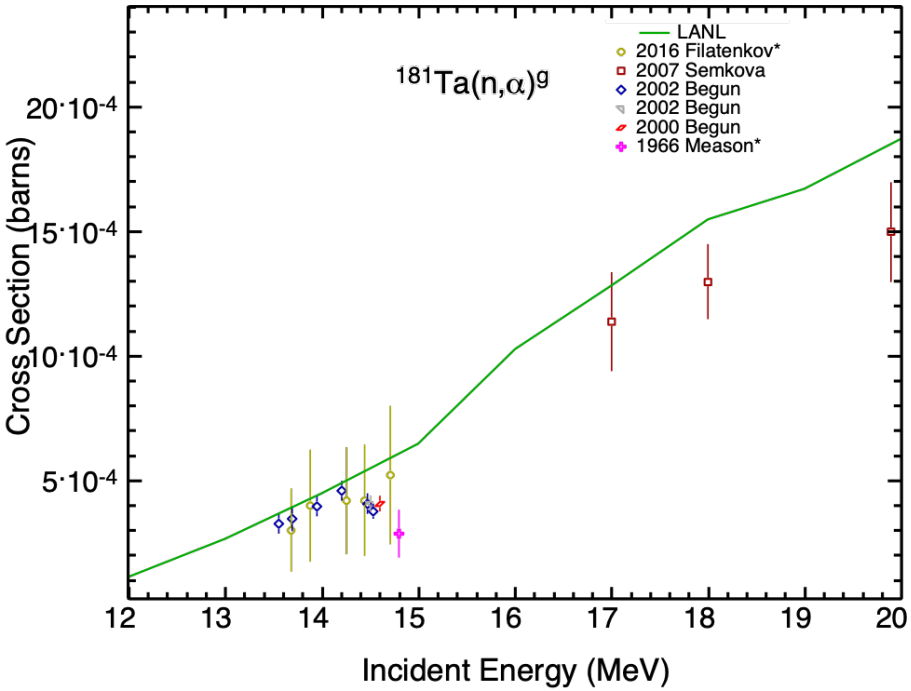
(n,p) evaluation was guided by Filatenkov's and by Semkova's data.

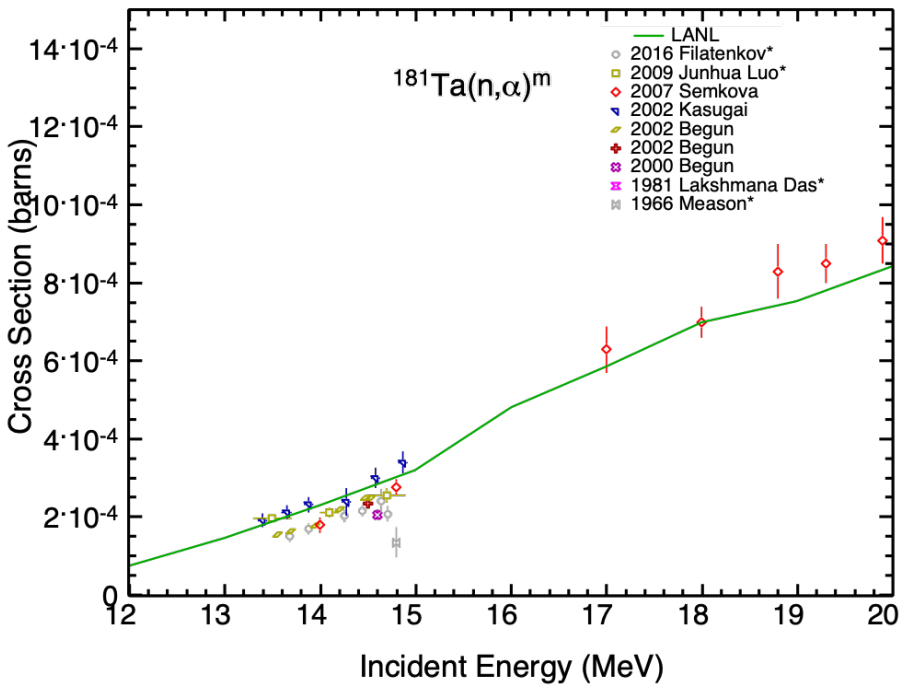
(n,α) reaction



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

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