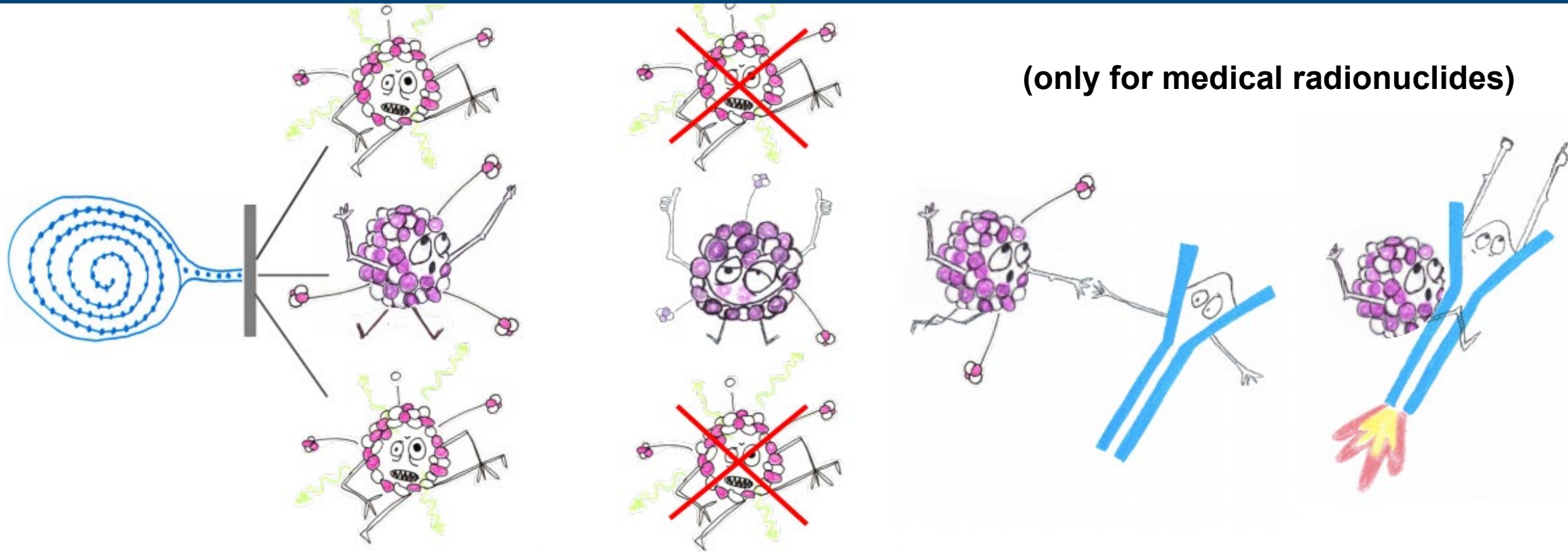


Investigating High-Energy
Proton-Induced Reactions:
Implications for Level Densities and
the Preequilibrium Exciton Model

Isotope Production (in a nutshell)



We go after the “holes” where experimental nuclear data do not exist by performing *targeted measurements* where there is a community-identified need!

4) Delivery

Needs:

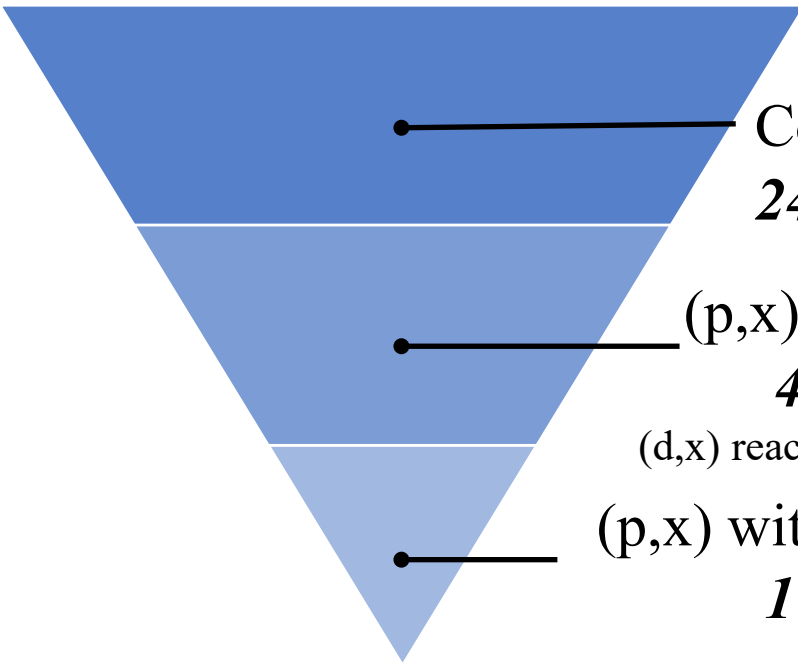
1

Tr

Cross sections

Decay data

The Nuclear Data Pipeline



Compilation (EXFOR)

24 000 datasets (46% for (n,x))

(p,x) reactions - 20%

4600 datasets

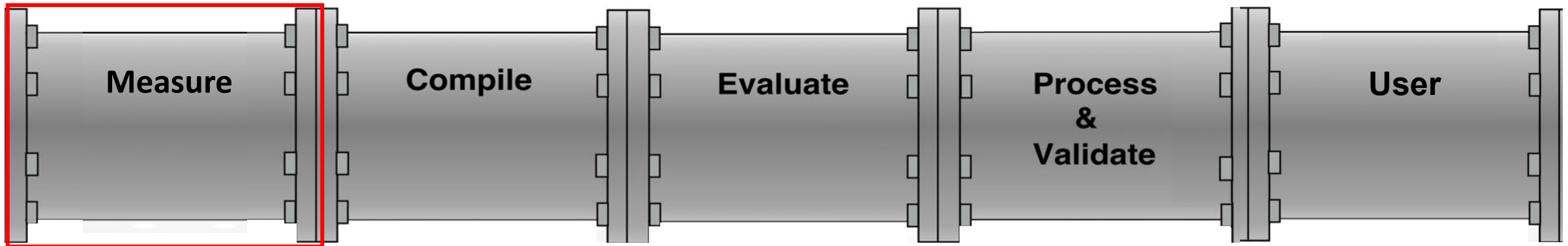
(d,x) reactions **2000** datasets – 9%

(p,x) with $E_p > 100$ MeV

10 datasets

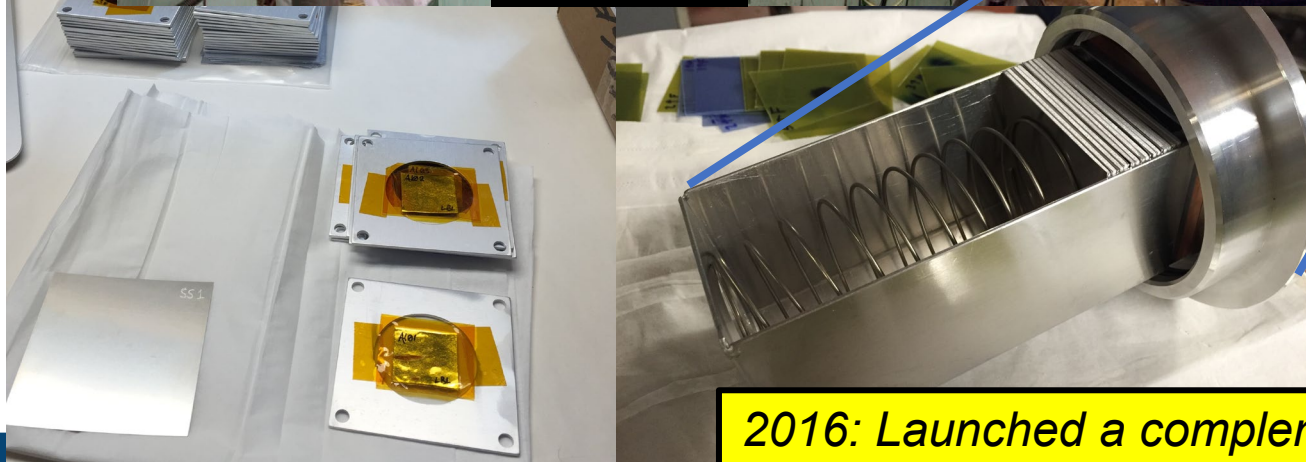
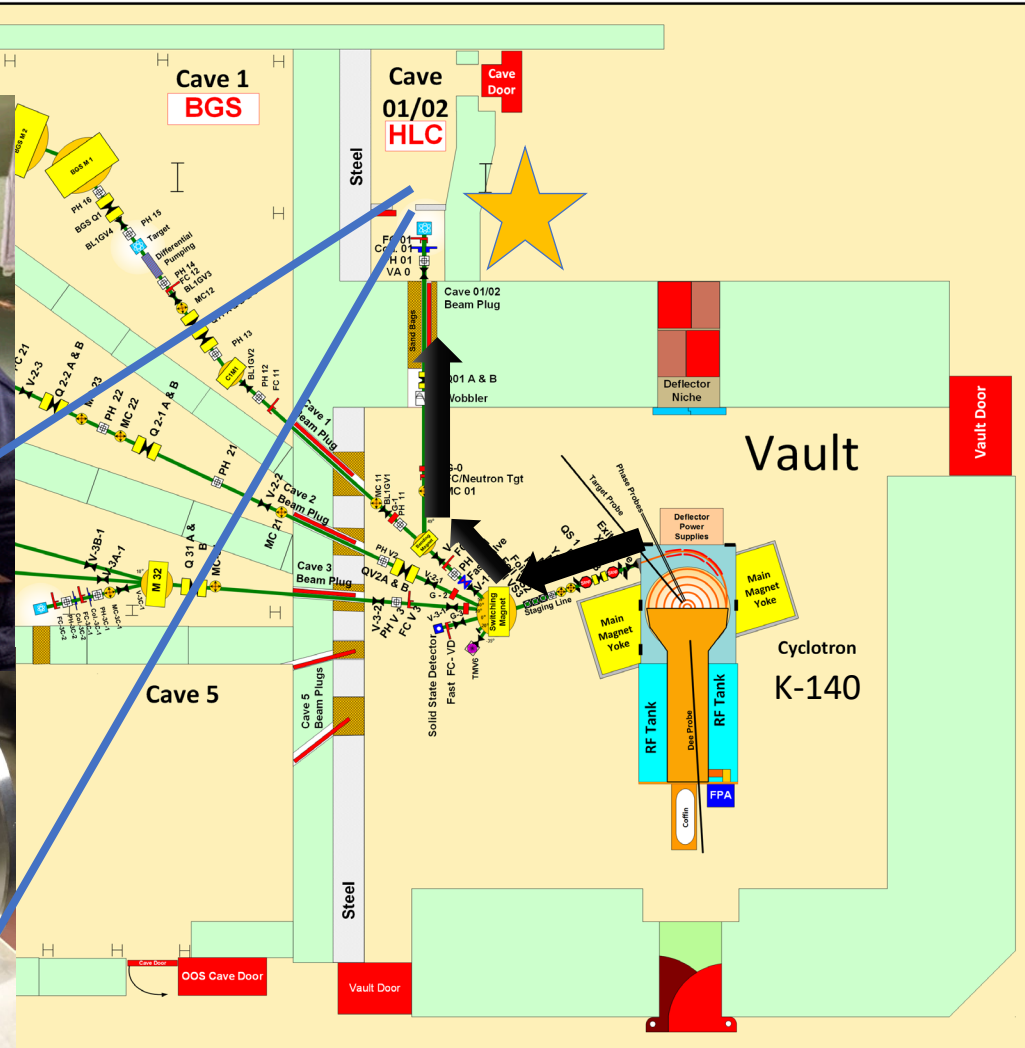
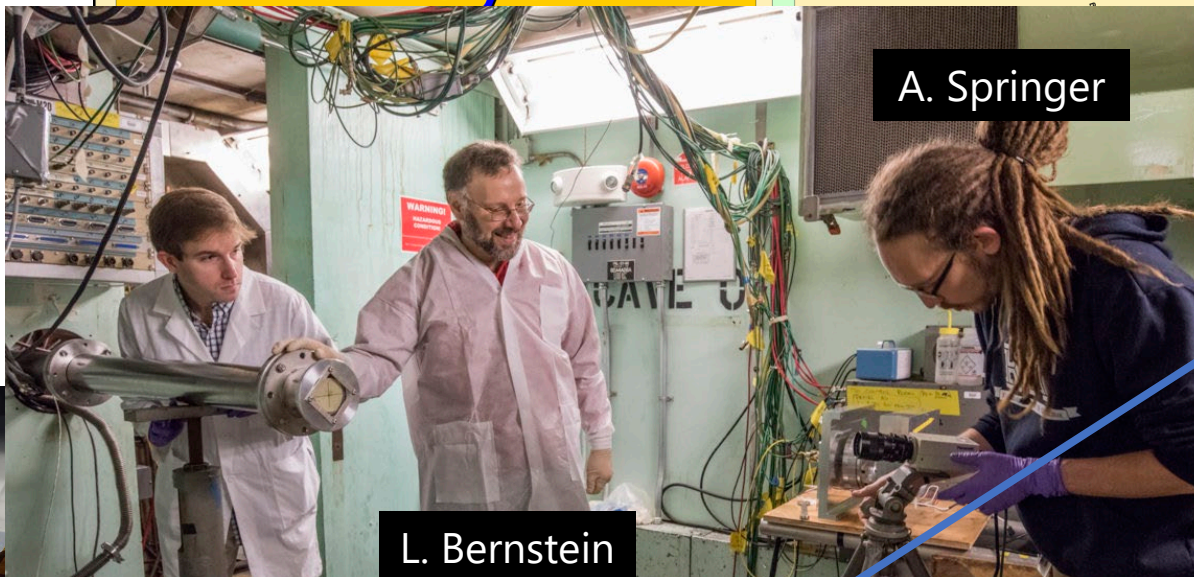
ENDF/BV-11.1: Neutron Reactions

0	1																	2													
n	H																	He													
	3	4											5	6	7	8	9	10													
	Li	Be											B	C	N	O	F	Ne													
	11	12											13	14	15	16	17	18													
	Na	Mg											Al	Si	P	S	Cl	Ar													
	19	20	21	22	23	24	25	26	27	28	29	30	31	32	33	34	35	36													
	K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr													
	37	38	39	40	41	42	43	44	45	46	47	48	49	50	51	52	53	54													
	Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe													
	55	56	57	72	73	74	75	76	77	78	79	80	81	82	83	84	85	86													
	Cs	Ba	La	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn													
	87	88	89	104	105	106	107	108	109	110	111																				
	Fr	Ra	Ac	Rf	Db	Sg	Bh	Hs	Mt	Ds	Rg																				
																	58	59	60	61	62	63	64	65	66	67	68	69	70	71	
																	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	
																	90	91	92	93	94	95	96	97	98	99	100	101	102	103	
																	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr	



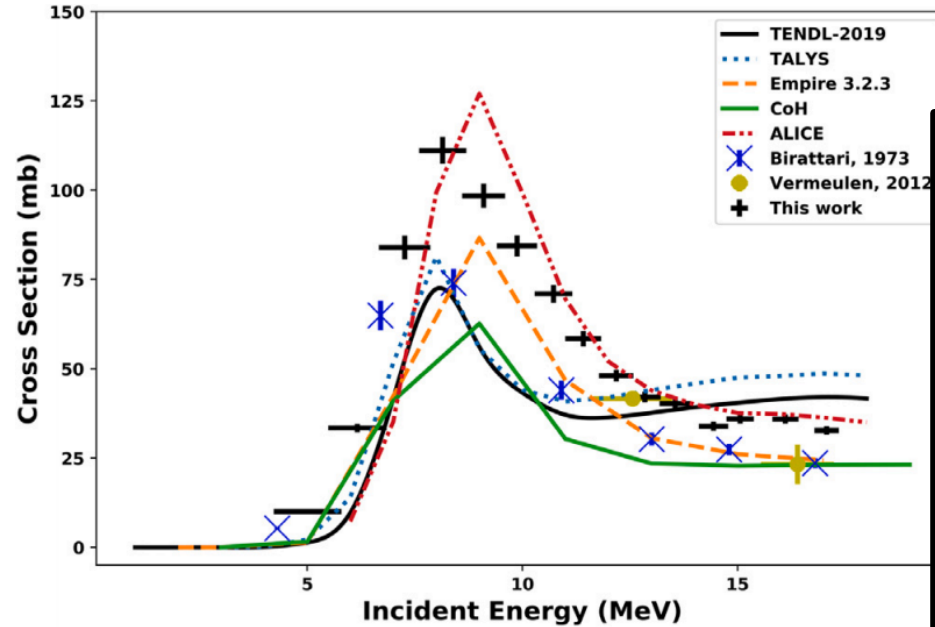
Stacked-Target Cross Section Measurements

88-Inch Cyclotron



2016: Launched a complementary nuclear data measurement campaign at the LBNL 88-Inch Cyclotron to address gaps in reaction data at $E < 60$ MeV/A

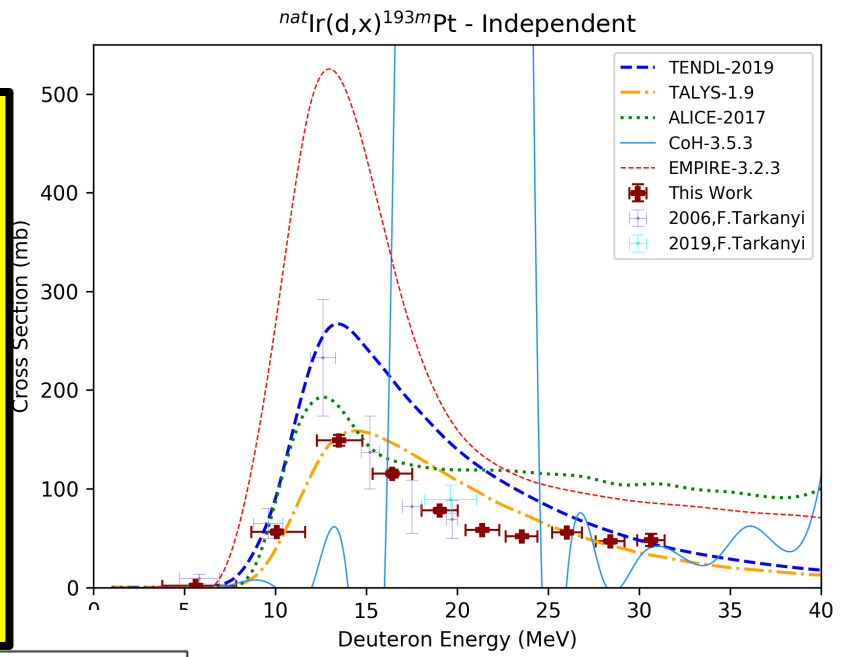
A Few Recent Highlights...



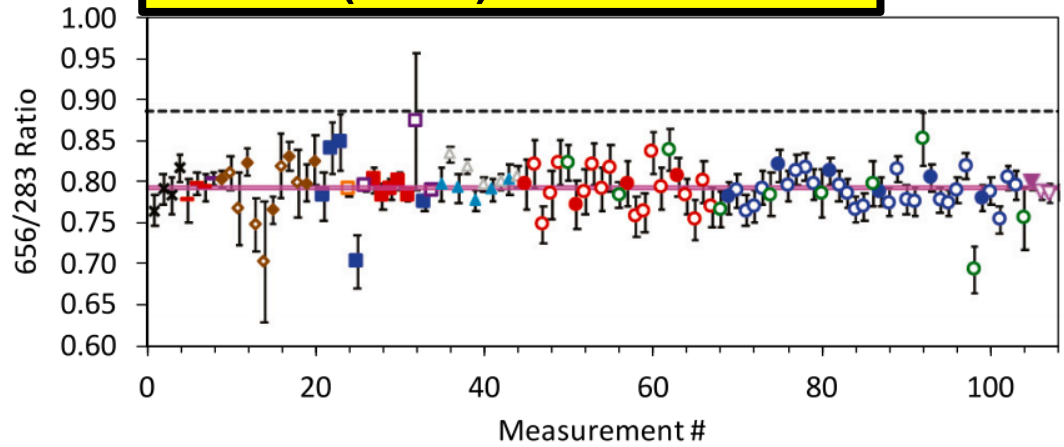
**Gd(p,x)¹⁶⁰Tb & the
“Tb Quartet”**

**Forthcoming: 4
pathways for ²²⁵Ac**

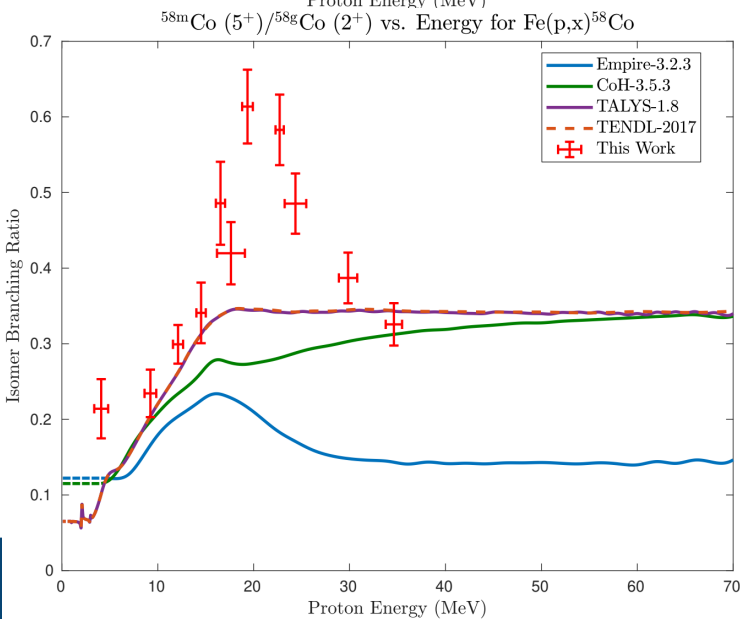
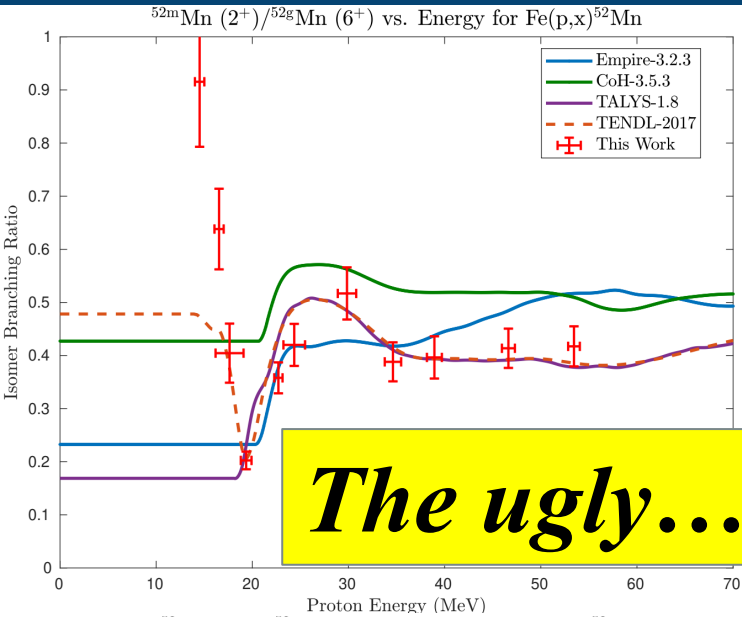
**Resolving γ intensity
discrepancies**
D.L. Bleuel, et al., *Applied
Radiation and Isotopes*,
170 (2021) 109625
M.S. Basunia, et al.,
Physical Review C, 101
(2020) 064619



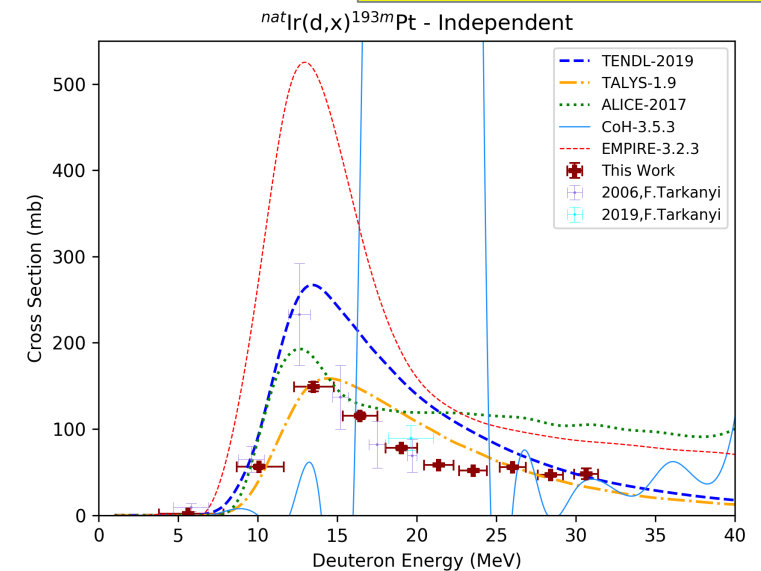
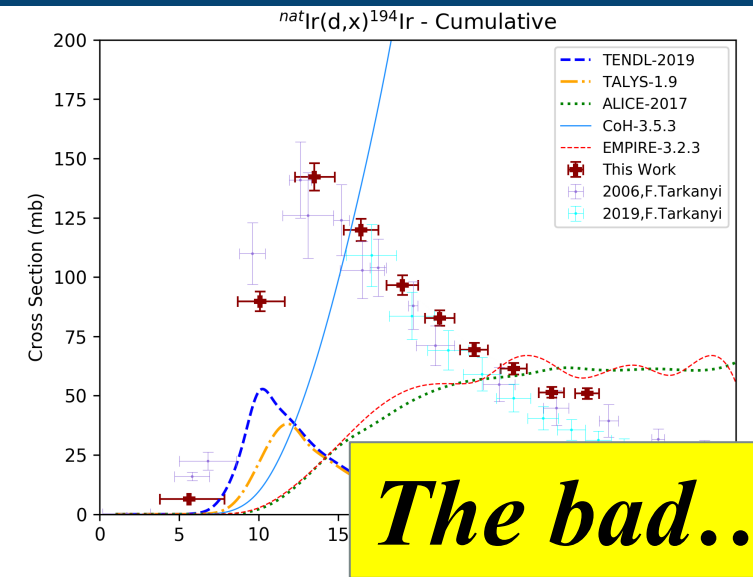
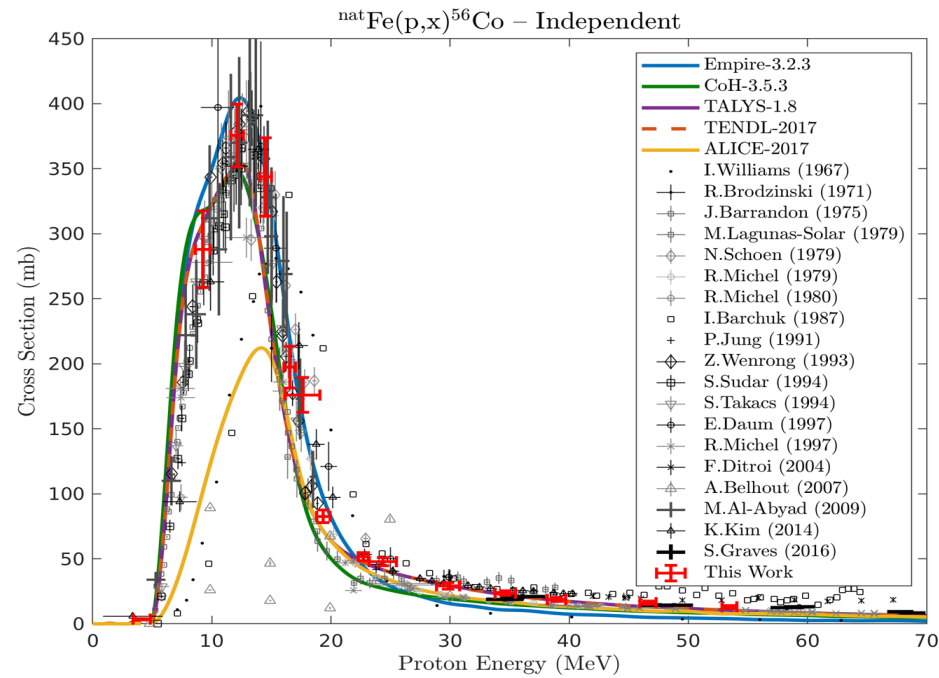
**Ir(d,x)^{193m}Pt for
targeted Meitner-
Auger therapy**



What If No Experimental Data Exist?



The good...



Isotope Production Research

A Tri-lab collaboration has been formed between LBNL, LANL, and BNL to measure (p,x) reactions relevant to isotope production from threshold to 200 MeV *for primary isotopes of interest and their impurities.*



LBNL 88-Inch Cyclotron
 $E_{p,max} = 60 \text{ MeV}$



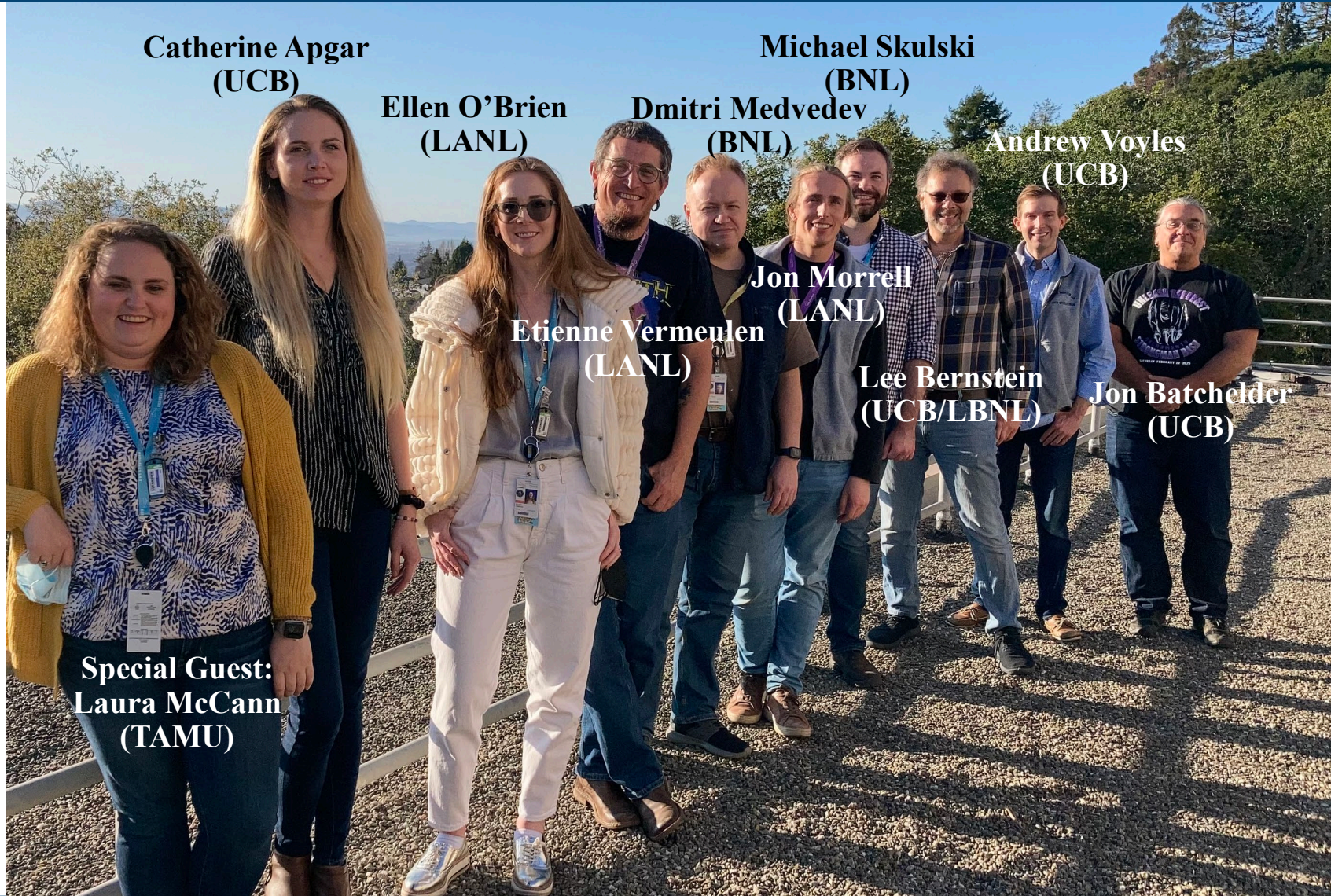
LANL IPF
 $E_{p,max} = 100 \text{ MeV}$



BNL BLIP
 $E_{p,max} = 200 \text{ MeV}$

The unique strength of this group is its access to all the different irradiations facilities and expertise

The Tri-Lab Effort in Nuclear Data



**Catherine Apgar
(UCB)**

**Ellen O'Brien
(LANL)**

**Michael Skulski
(BNL)**

**Dmitri Medvedev
(BNL)**

**Andrew Voyles
(UCB)**

**Jon Morrell
(LANL)**

**Etienne Vermeulen
(LANL)**

**Lee Bernstein
(UCB/LBNL)**

**Jon Batchelder
(UCB)**

**Special Guest:
Laura McCann
(TAMU)**

Not Pictured:

Eva Birnbaum (LANL)

Cathy Cutler (BNL)

Amanda Lewis (UCB)

Arjan Koning (IAEA)

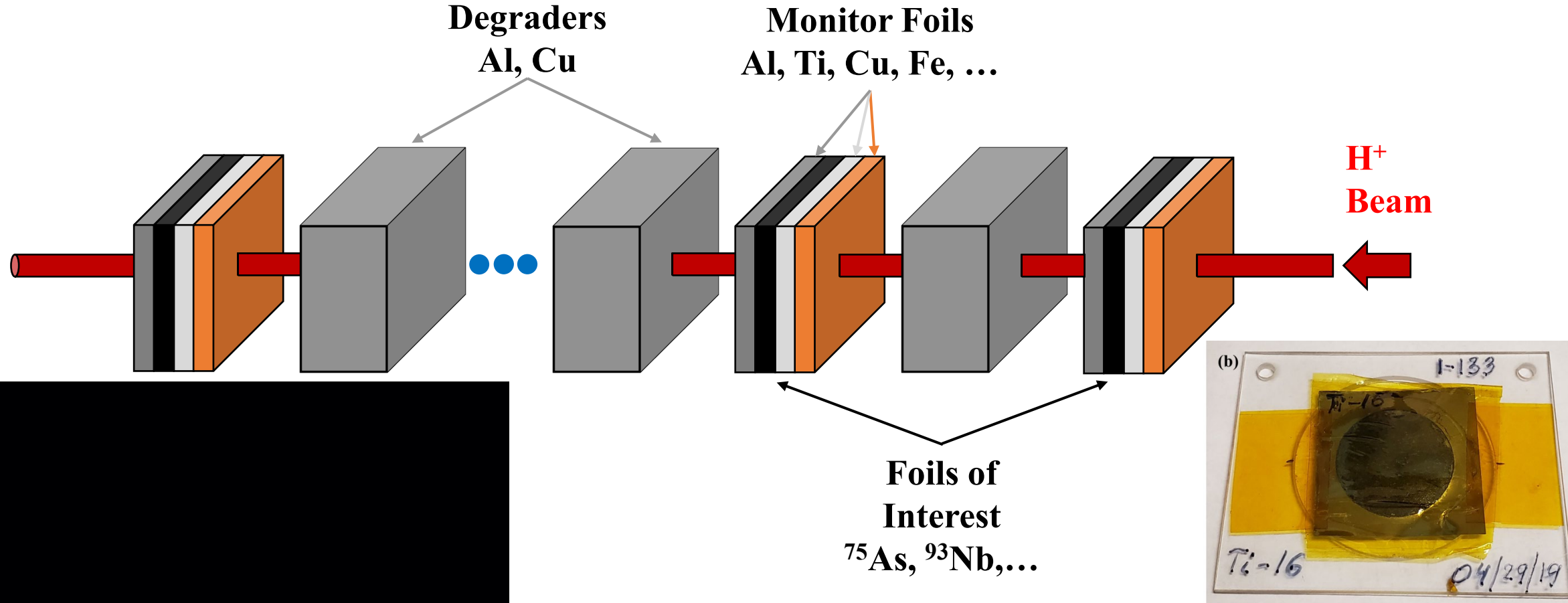


**Meiring Nortier
(LANL)**



**Morgan Fox
(UCB)**

Stacked-Target Experimental Method



Check out Curie! (and help improve it!)

<https://jtmorrell.github.io/curie/build/html/index.html>

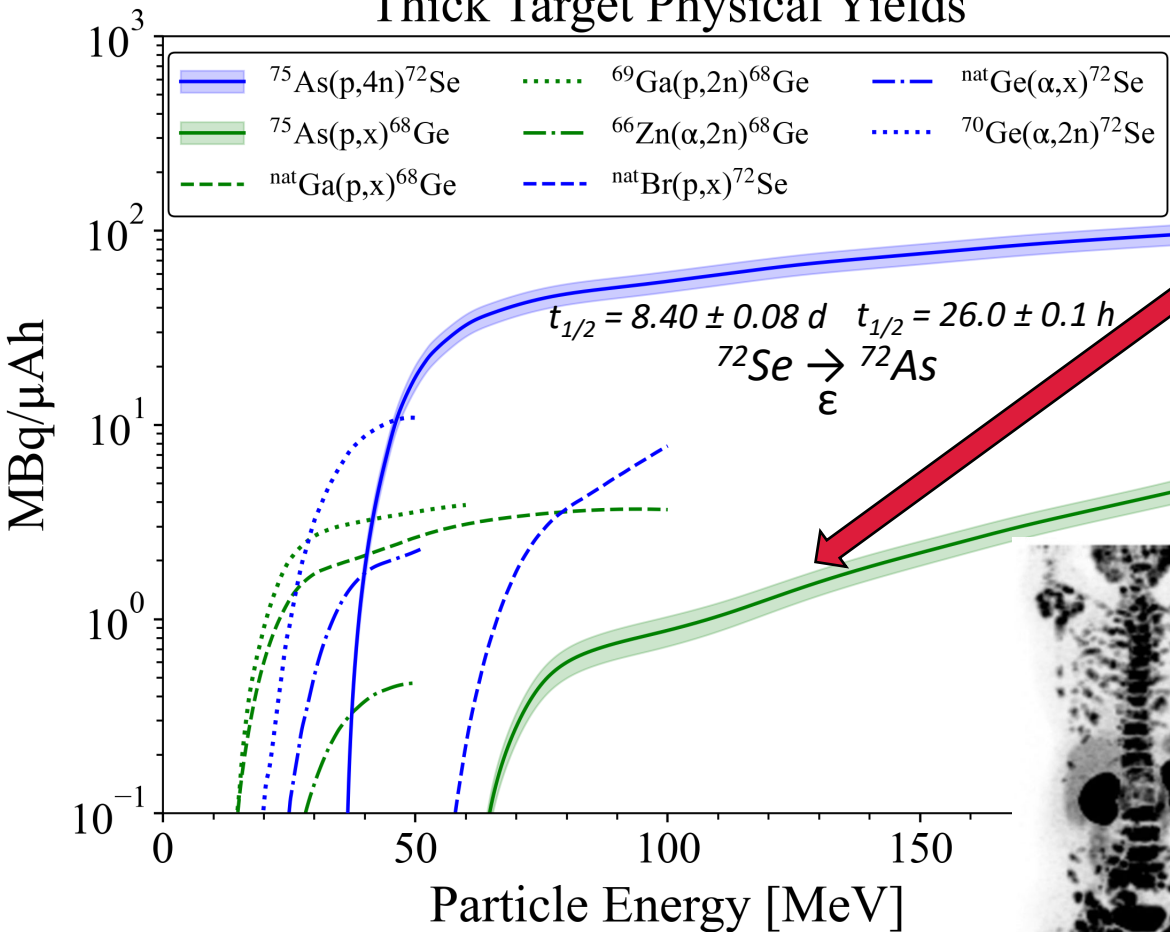
A.S. Voyles et al., *NIM B* 429 (2018) & *EPJ A* 57:94 (2021)

J.T. Morrell et al., *EPJ A* 56:13 (2020)

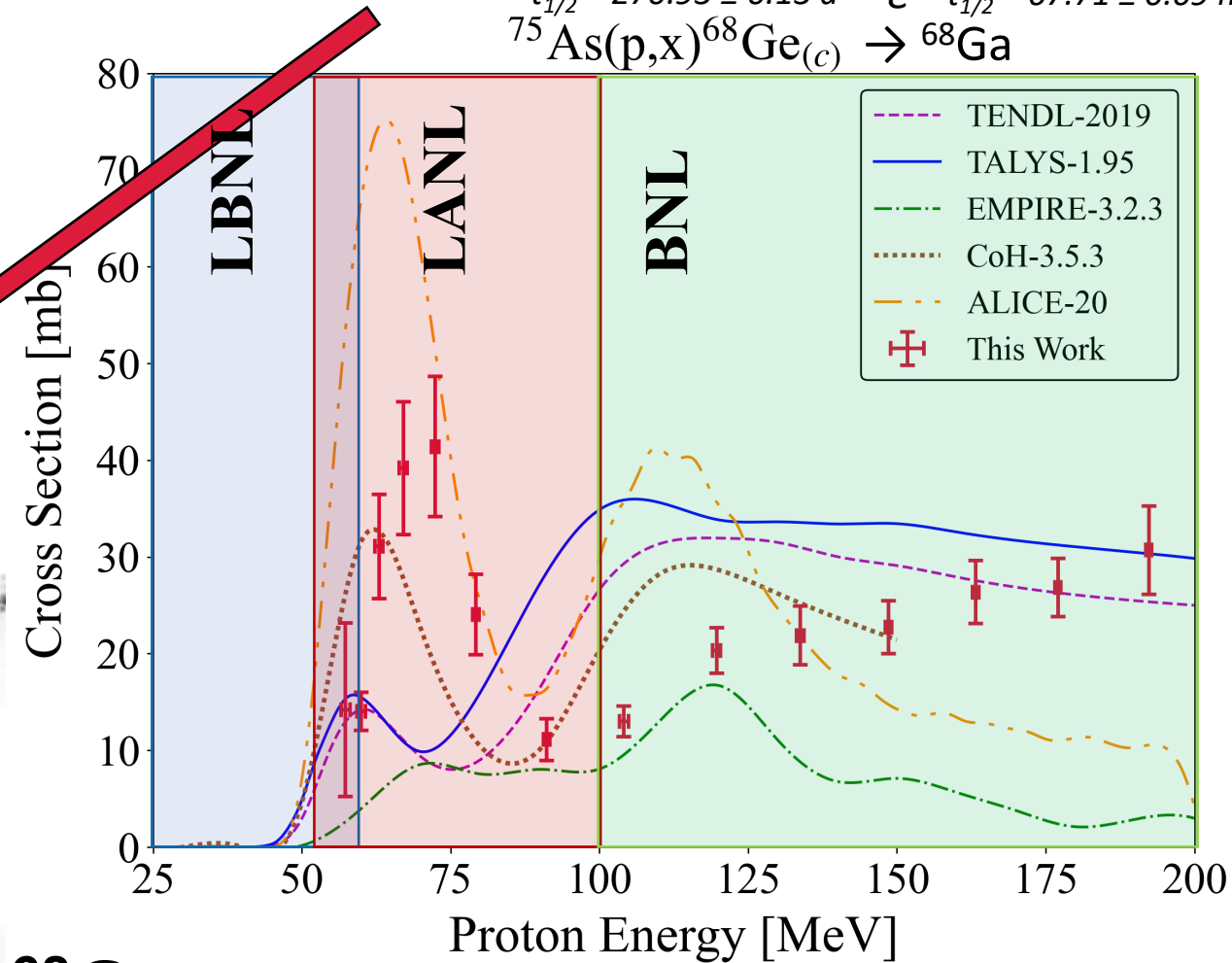
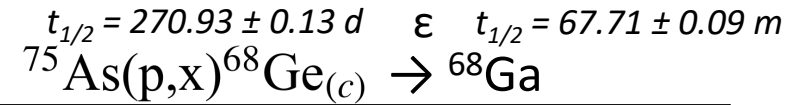
M. B. Fox et al., *PRC*, 103(3):034601 (2021) & *PRC* 104, 064615 (2021)

^{72}Se and ^{68}Ge Production

Thick Target Physical Yields



Arsenic route offers highest yield and purity for ^{72}Se

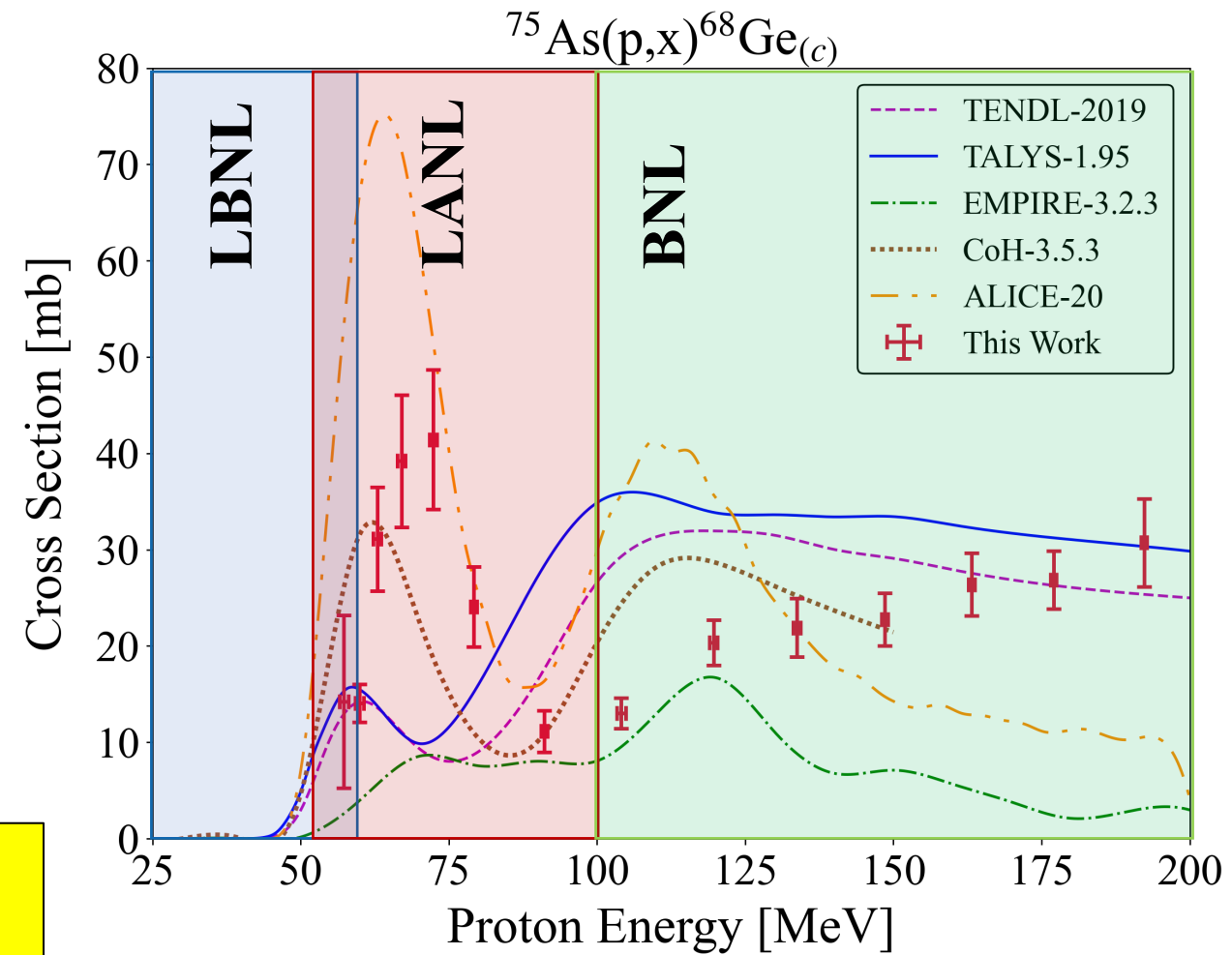


^{68}Ga

Nuclear Data Contributions – Medical and Models

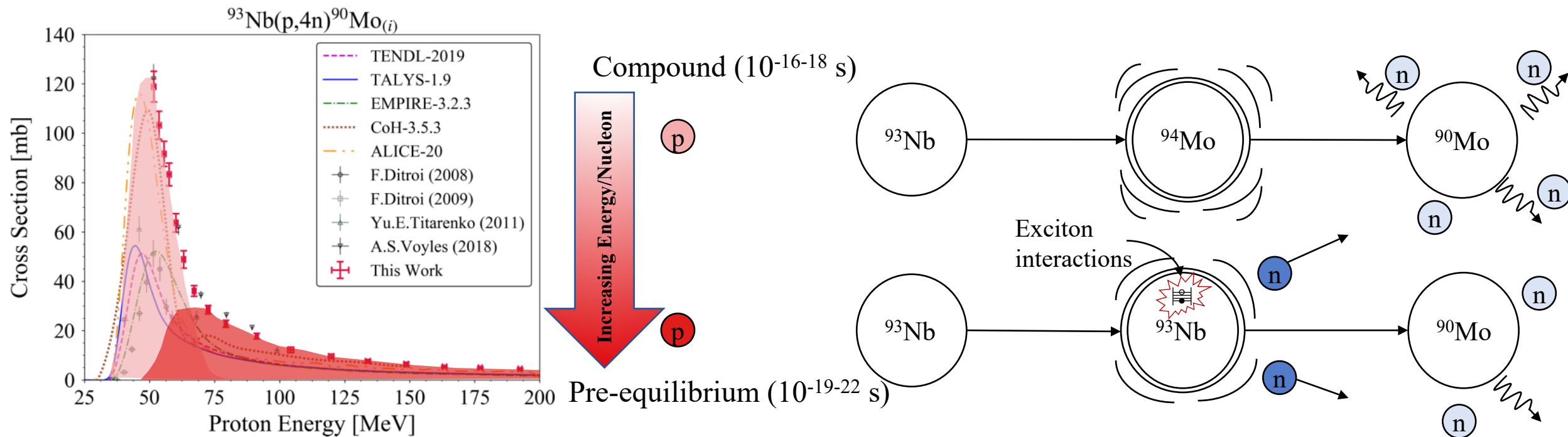
- These data add direct value for isotope production
- But we are also a nuclear data group
 - This is a large & self-consistent contribution of scarce data!
 - A valuable opportunity to study high-energy reaction modeling and evaluation

How do we improve modeling for high-energy proton-induced reactions?

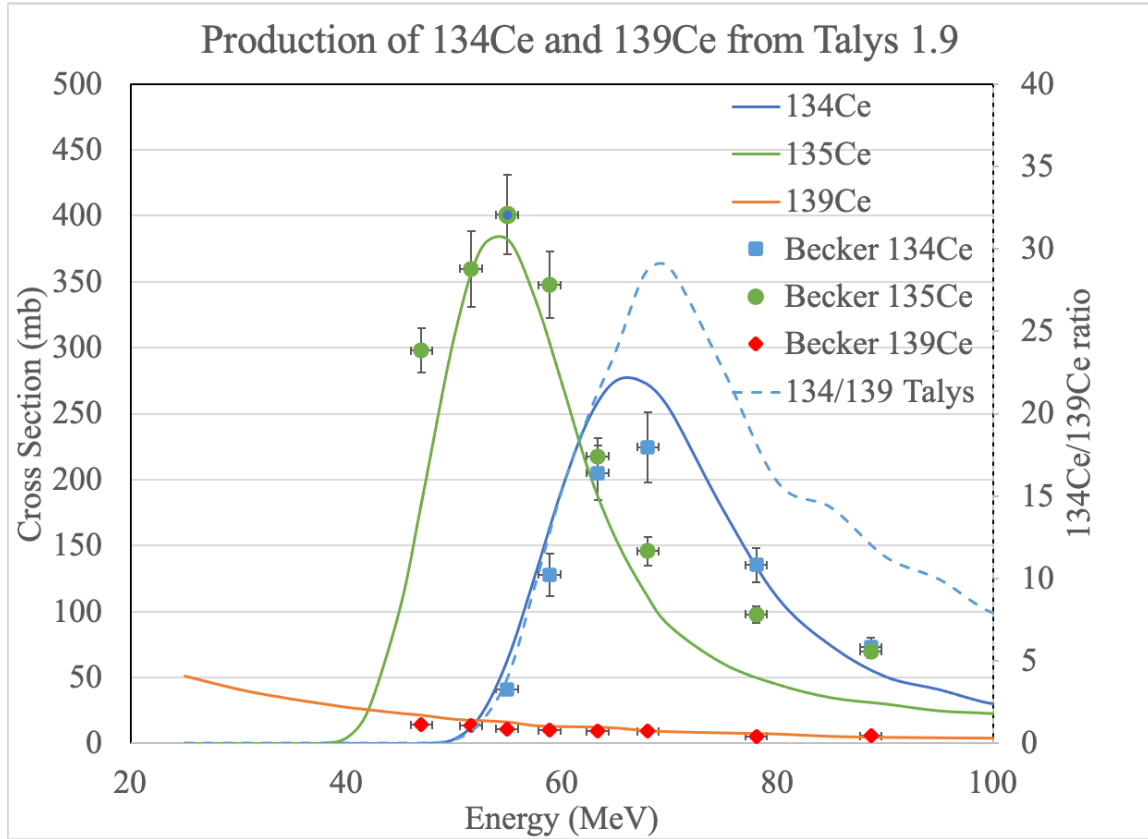


Impact Beyond “Better Fitting Lines”

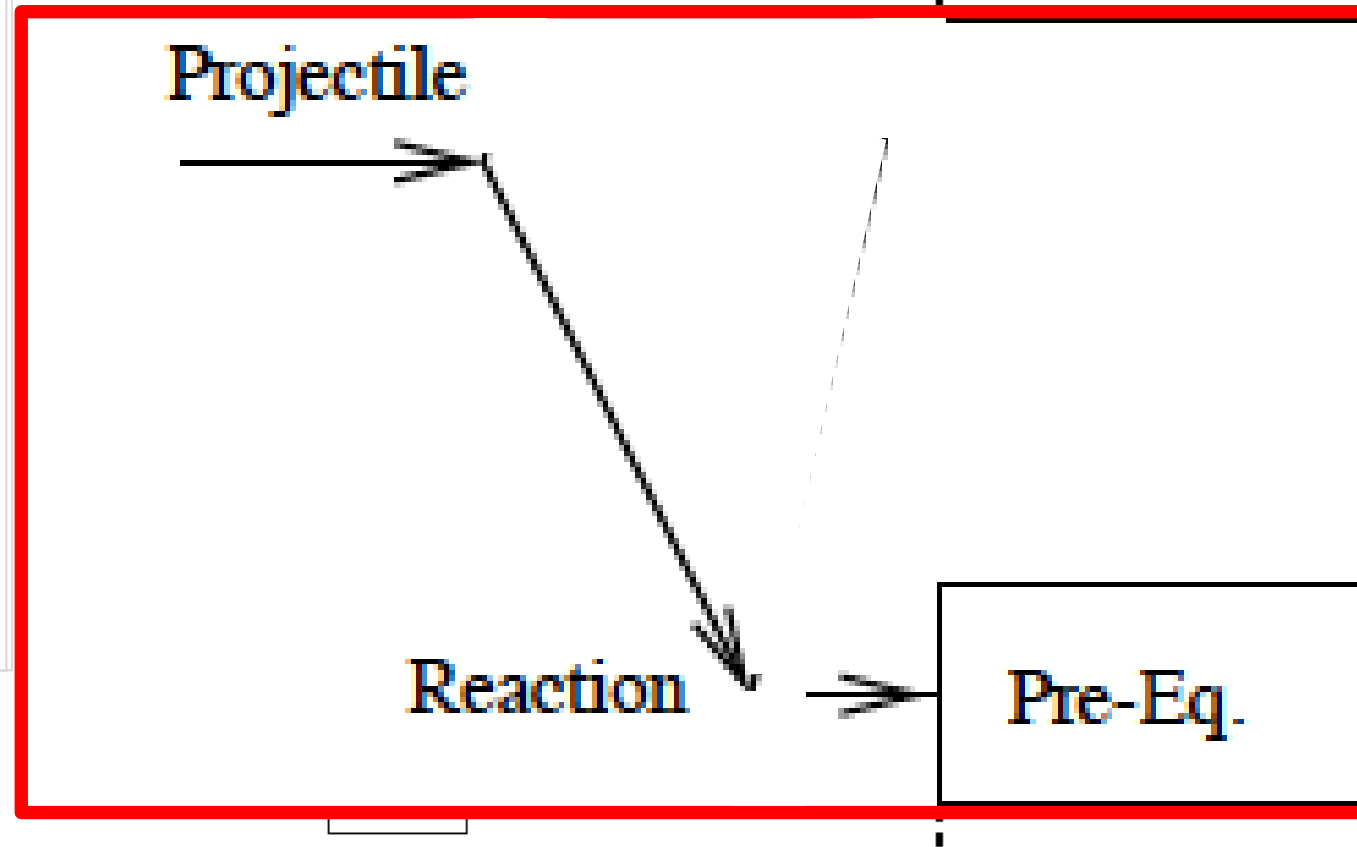
- The goal is not surface level improvements in χ^2
 - The reflection of physics adjustments made and the process are vitally important pieces
- This is the start to an evaluation approach and the comments we make are substantial because they immediately become a basis for these regimes



The TREND experimental effort* seeks an improvement in the ability of reaction modeling to optimize radioisotope production



But there are literally ***thousands*** of other parameters that can be adjusted!

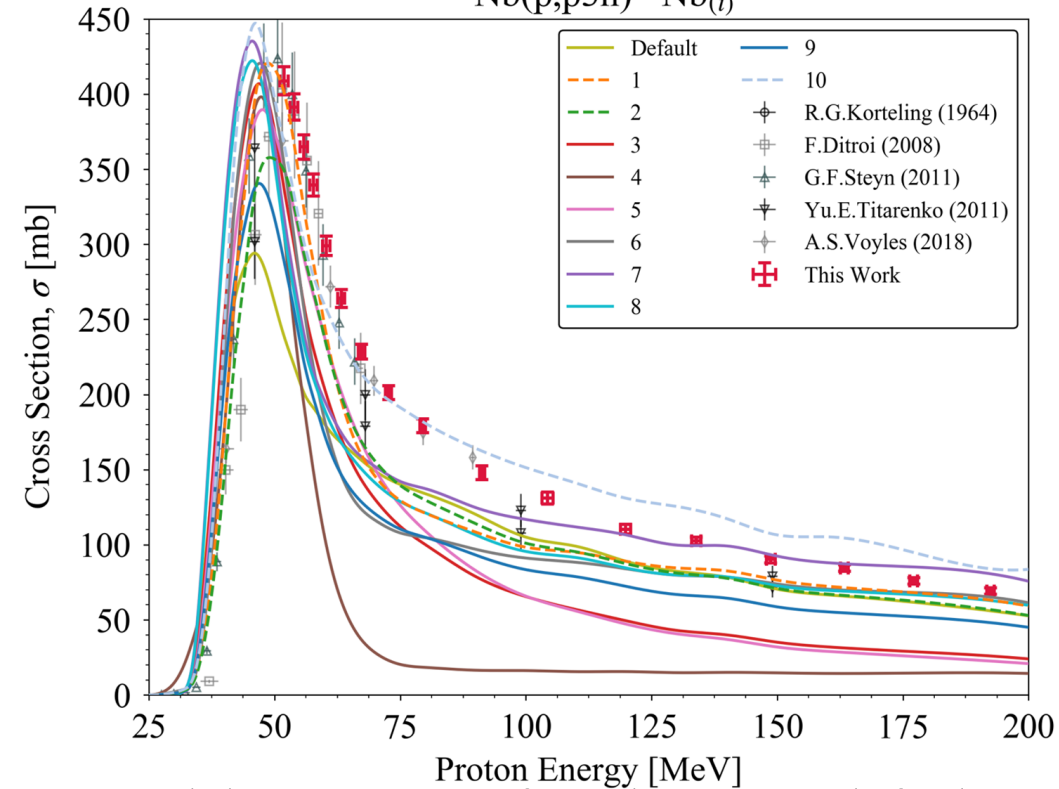
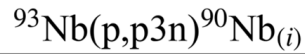


More data are needed to obtain the correct set of reaction parameters

What Happens if We Don't Do the Right Thing?

- Consider 10 different models, with arbitrary choices of which simplistic or complex parameters are adjusted, to reproduce similar improvements over the default prediction...

What Happens if We Don't Do the Right Thing?



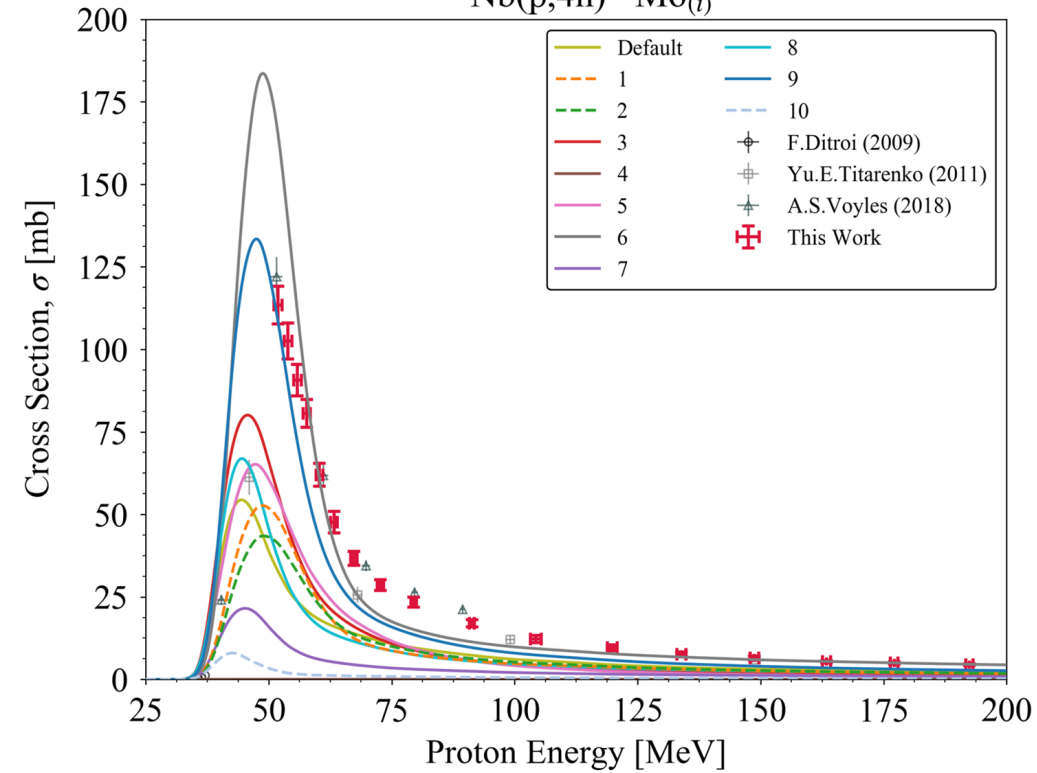
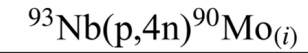
Models 1, 2, 10 perform best over default

with arbitrary cl
ted, to reproduce

, what parameter

ible
engh

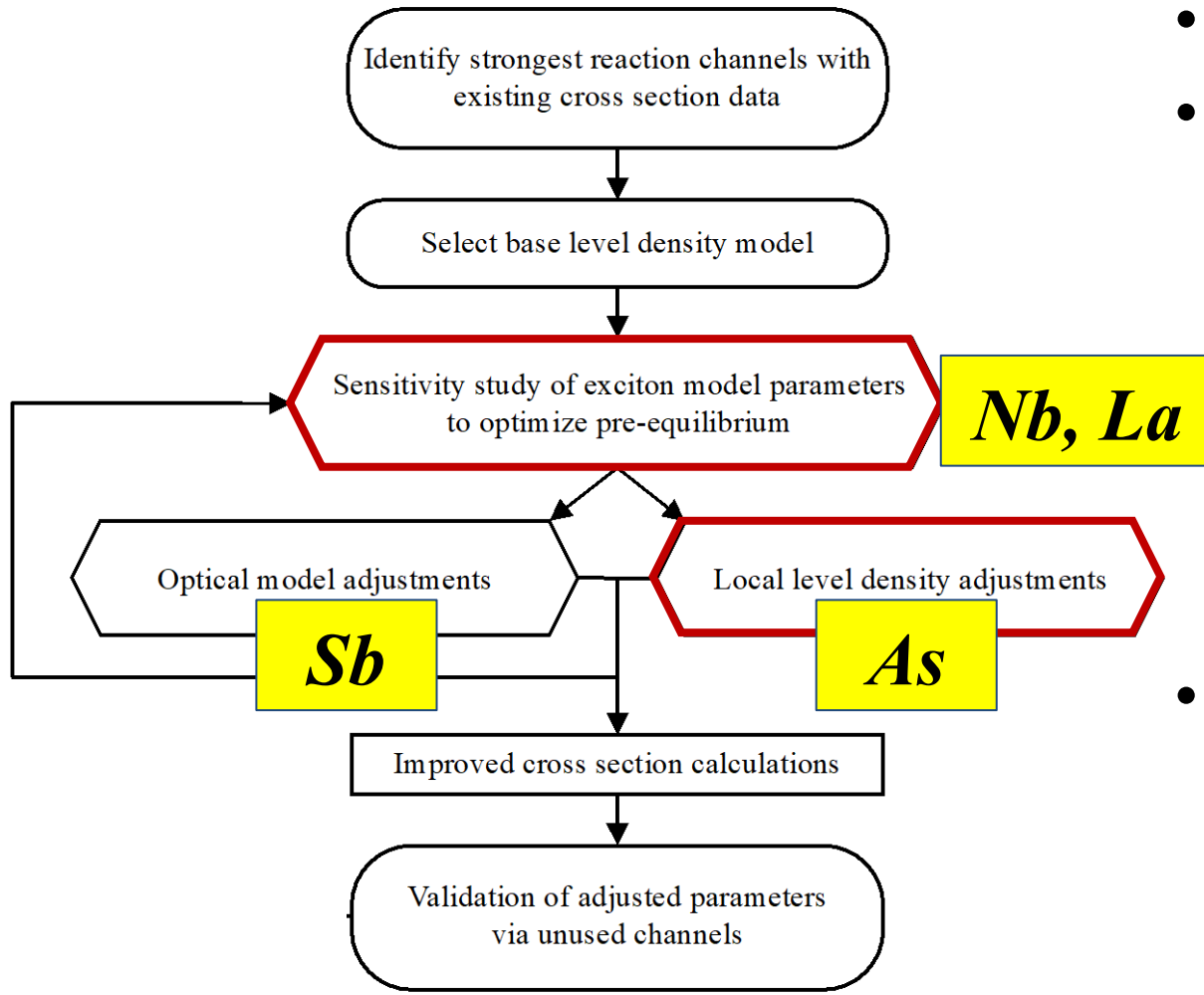
onstant, $M2shift$, $M2l$
 $w1adjust$, $v1adjust$



Models 1, 2, 10 perform extremely poorly

Single-channel optimizations lead to non-unique, non-physical solutions!

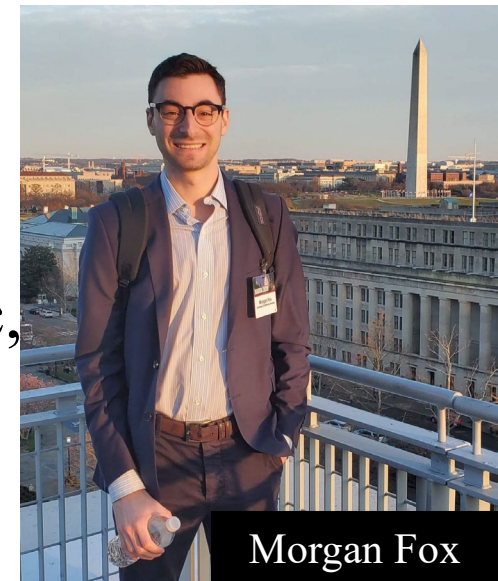
TREND results were used to develop a new data evaluation methodology for high-energy (p,x) reactions



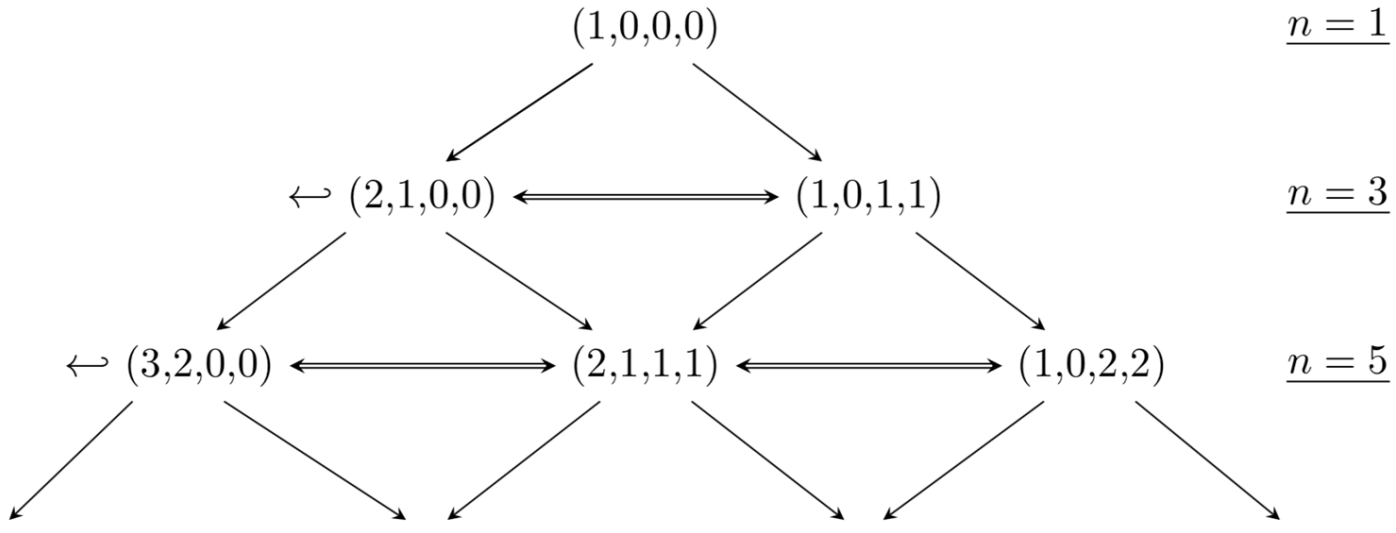
- Based in TALYS code
- We established a collaboration with the TALYS lead developer, Dr. Arjan Koning (Head of the IAEA Nuclear Data Section)

Emphasis placed on pre-equilibrium parameter adjustments to match the strongest-fed channels

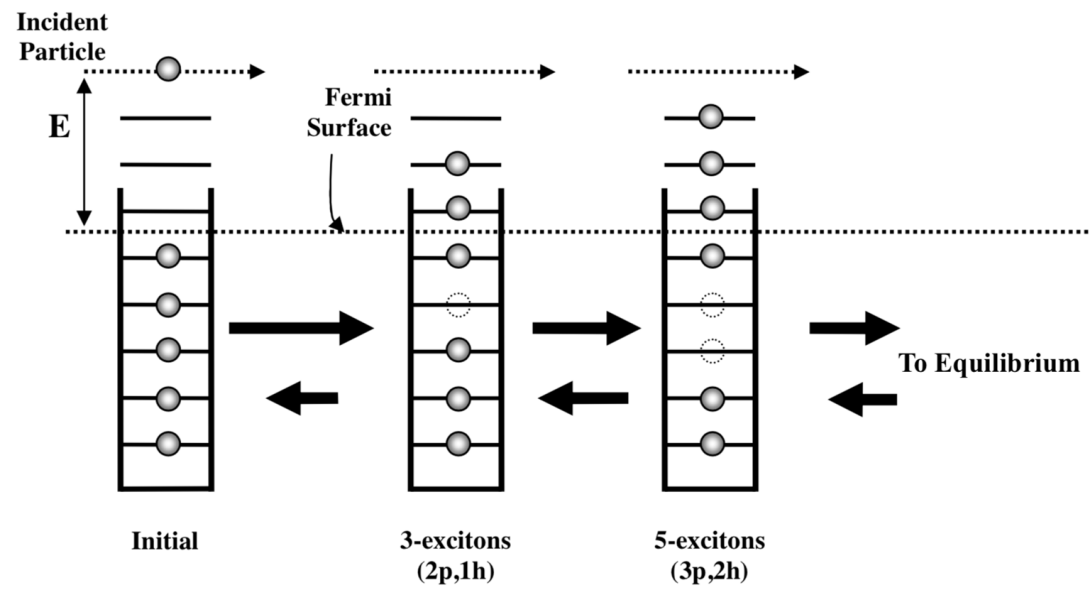
- The modeling is validated via comparison to nonelastic, cumulative channels (same product formed by several exit channels)



Methodology provides insight into pre-equilibrium reaction dynamics and a host of nuclear data properties relevant to accurate modeling



$n = 1$
 $n = 3$
 $n = 5$



$M2constant$ $M2limit$

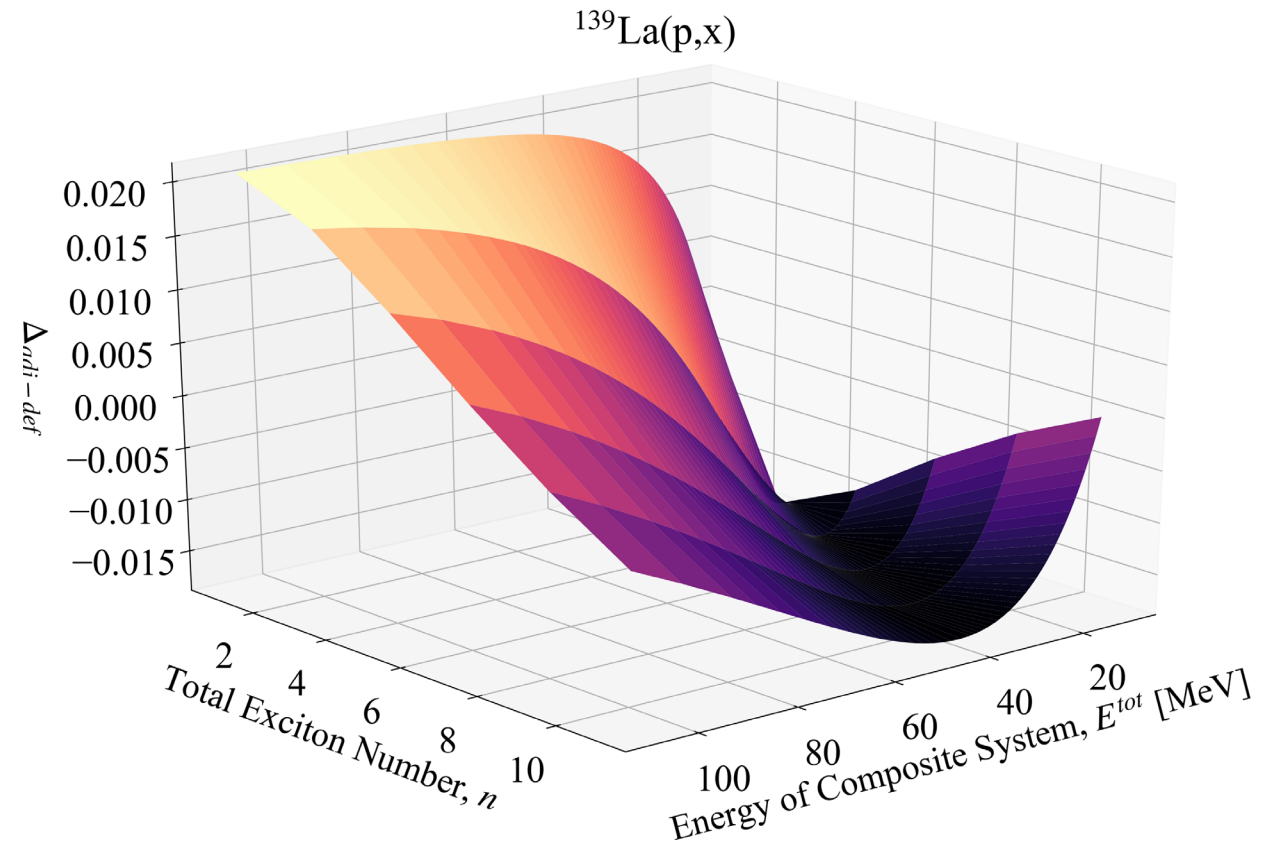
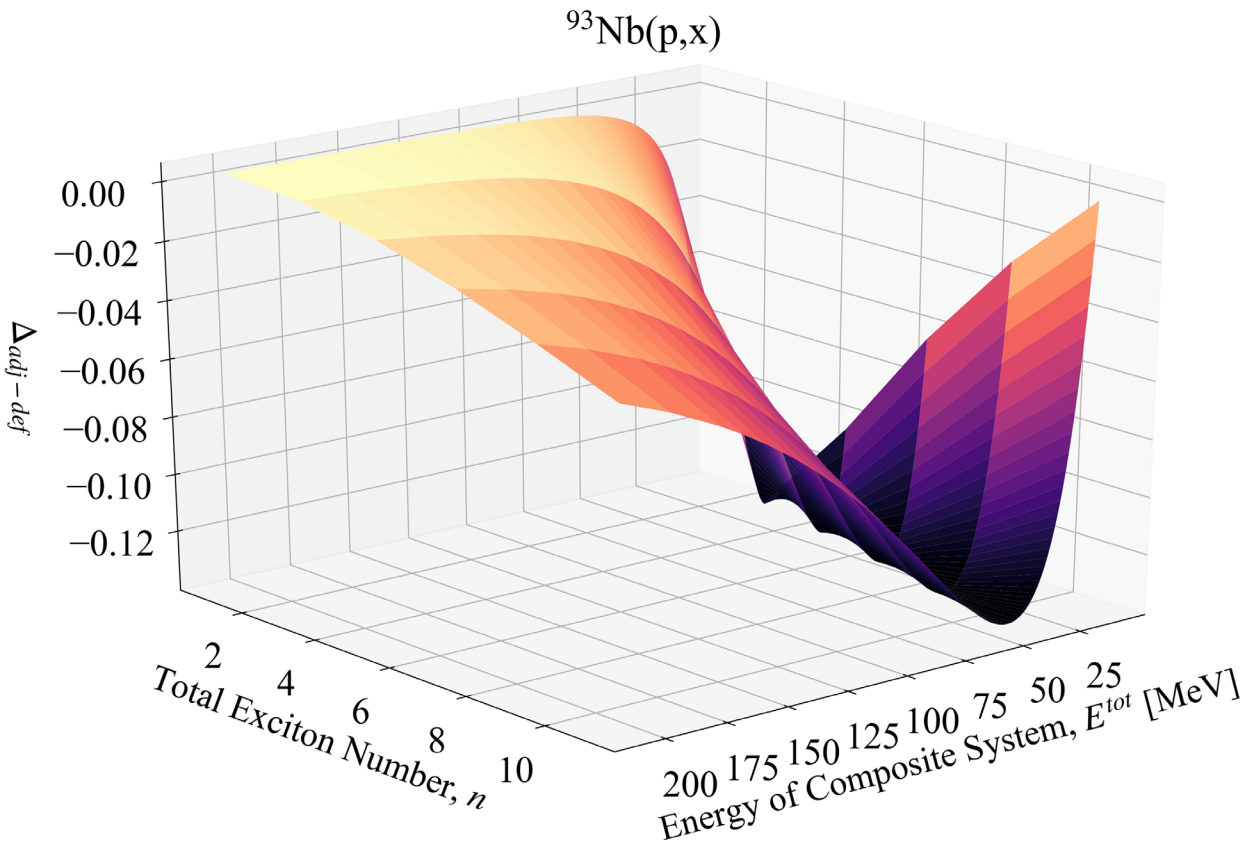
$$M^2 = \frac{C_1 A_p}{A^3} \left[7.48 C_2 + \frac{4.62 \times 10^5}{\left(\frac{E^{tot}}{n A_p} + 10.7 C_3\right)^3} \right]$$

$M2shift$

$$\Delta_{adj-def} = \frac{M^2(E^{tot}, n)_{adj}}{M^2(E^{tot}, n)_{adj,max}} - \frac{M^2(E^{tot}, n)_{def}}{M^2(E^{tot}, n)_{def,max}}$$

Exciton strength parameters are key for modeling pre-equilibrium emission!

Analyzing a Trend in Exciton Adjustments

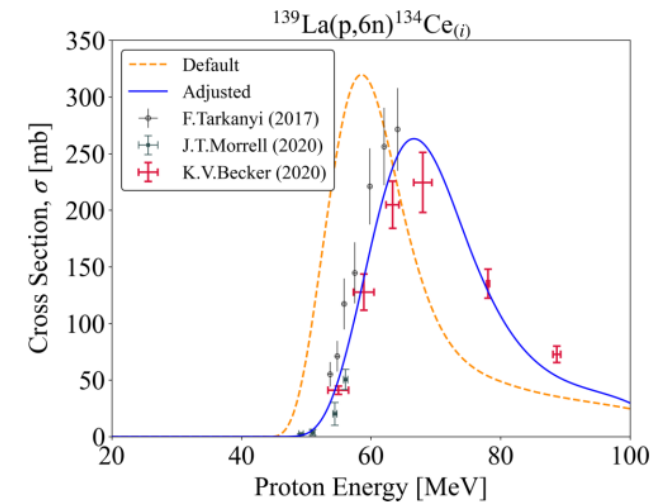
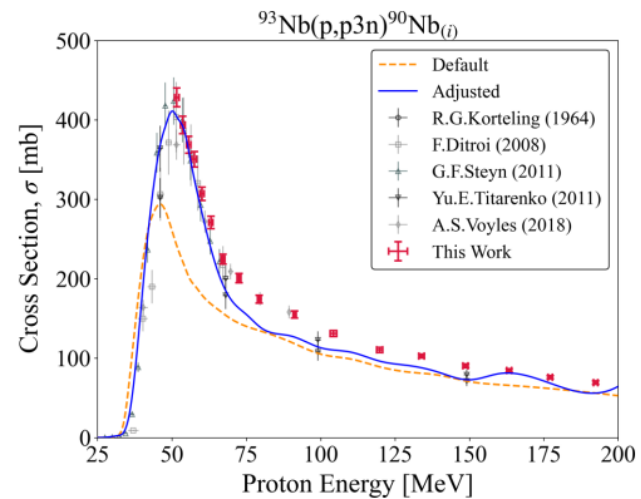
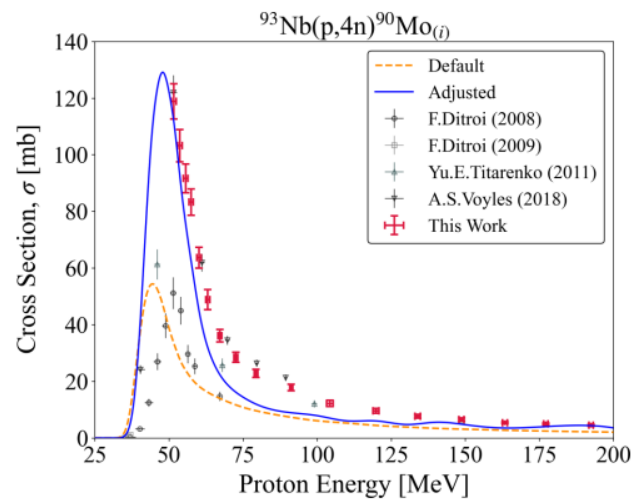


Significant differences are consistently seen in the energy region where we transition from the compound to pre-compound models!

Evaluation Procedure applied to $^{93}\text{Nb}(p,x)$, $^{139}\text{La}(p,x)$

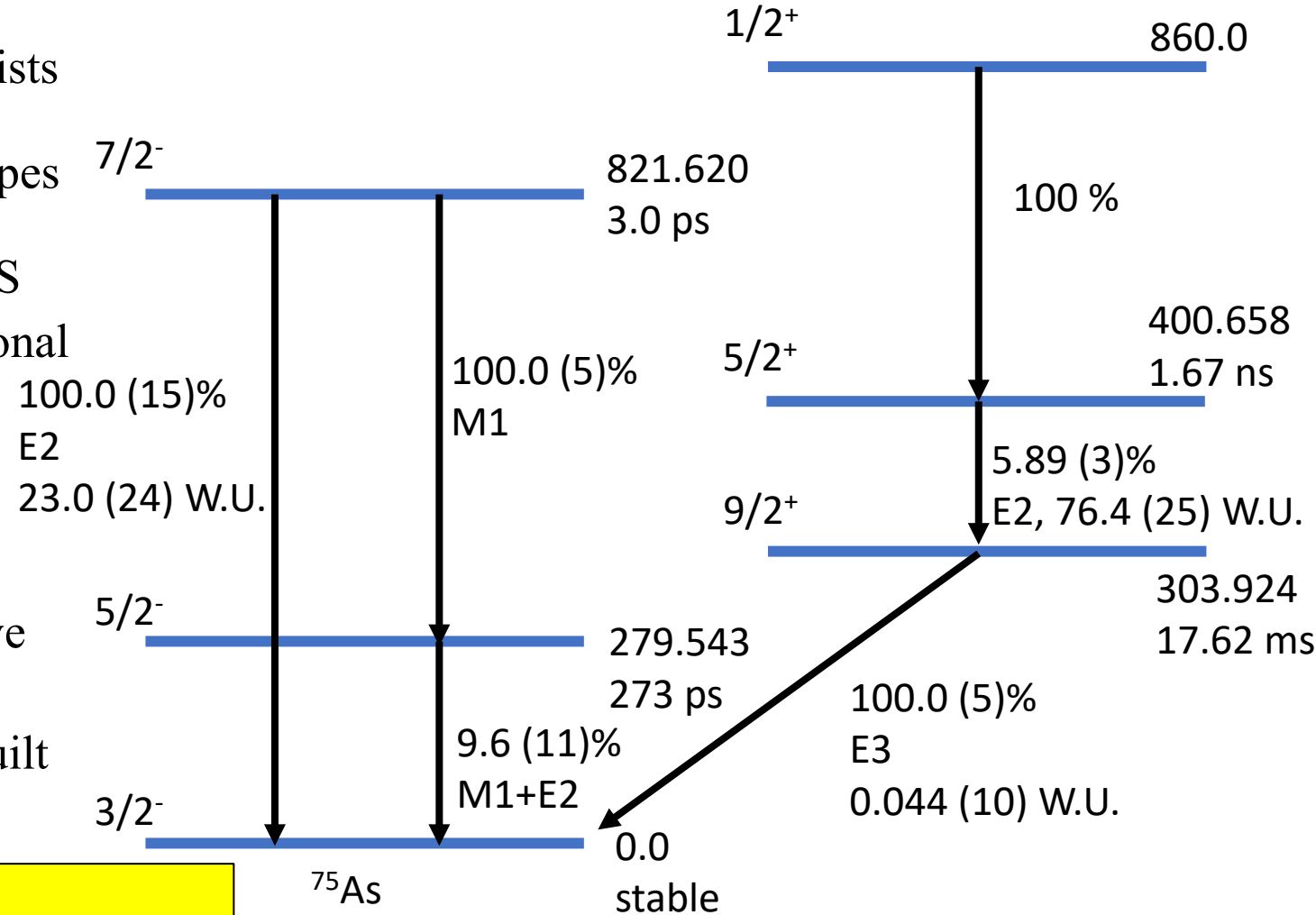
- Exciton model adjustments in this mass region have led to significant improvements in pre-equilibrium, with global χ^2 improvements up to 40x
- However, the base level density (**ldmodel 4**, Goriely HFB + Skyrme) needed to be changed (**ldmodel 5**, Hilaire HFB + Skyrme) for most Nb, Mo products

All is not well in the state of Denmark w.r.t. level density!



Level Density Adjustments in $^{75}\text{As}(p,x)$

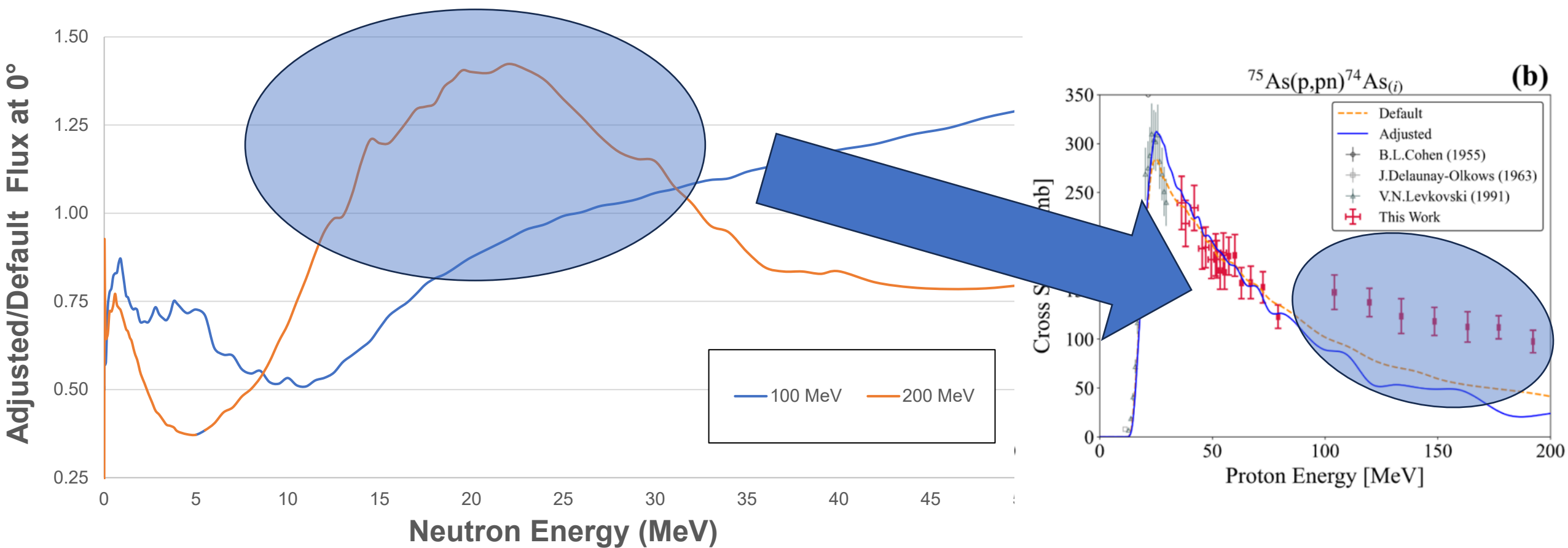
- RIPL-3 suggests an oblate deformation of $\beta_2 = -0.25$
- Nilsson diagram systematics for neutron-rich isotopes near $A=60-80$ indicate prolate, ENDF lists $\beta_2 = 0.314(6)$
- TALYS lacks deformation coupling for As isotopes and does a spherical OMP calculation via ECIS
- 3-level apparent rotational band added to TALYS
- Neighboring $^{76,74}\text{Se}$, $^{76,74}\text{Ge}$ demonstrate vibrational character, have vibrational coupling schemes in TALYS for CC
- ^{241}Am is only odd-Z nucleus with vibrational deformation in TALYS
- Since TALYS ECIS is unsuited for purely vibrational coupling schemes in odd-Z nuclei, we were forced to use weak-coupling model and modeled ^{75}As as soft vibrational, with a weak vibrational band (based on ^{241}Am formatting) built on top of the G.S. rotational band



TALYS also lacks isospin....

*M. B. Fox *et al.*, *PRC*, 103(3):034601, 2021 & *PRC* 104, 064615 (2021) ²²

What are the effects of these changes on the neutron flux look like behind a thick target at BNL-BLIP or LANL-IPF?



There is a sizable neutron flux behind all thick target stacks at these Isotope Production facilities!

Level Density Adjustments in $^{75}\text{As}(p,x)$

- Like with Nb/La, we see a clear need to transition to a new level density model as we get far from target

- Effect is more prominent in As, as we produce isotopes further from stability
- More pronounced for (p,xn) channels
- This does NOT imply that one ldmodel is better than others – *none work globally for high E_x !*

This is a problem!

A new Gogny + QRPA “ldmodel 7”?

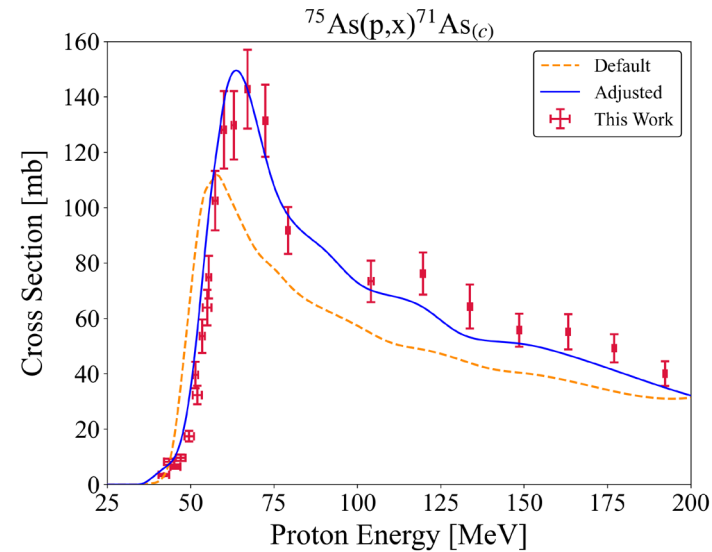
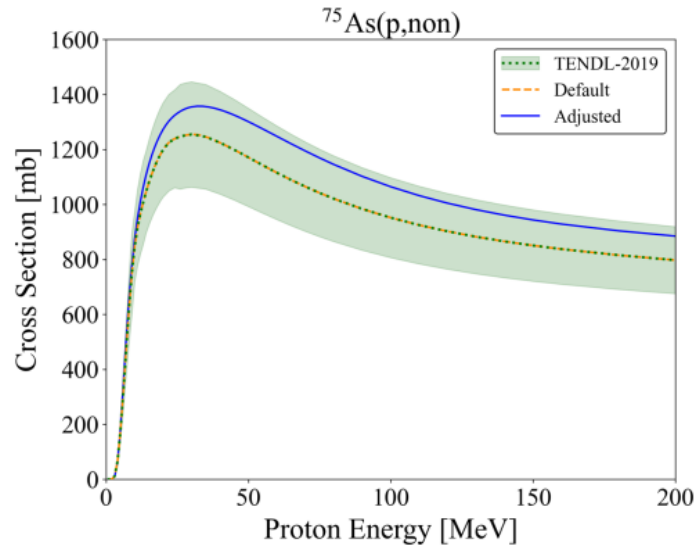
→ **ldmodel 4**, Goriely HFB + Skyrme
 → **ldmodel 5**, Hilaire HFB + Skyrme

All others → **ldmodel 6**, Temperature-Dependent HFB + Gogny

34	^{64}Se 33 ms	^{65}Se 33 ms	^{66}Se 133 ms	^{67}Se 35.5 s	^{68}Se 27.4 s	^{69}Se 41.1 m	^{70}Se 4.74 m	^{71}Se 8.40 d	^{72}Se 38.8 m, 7.15 h	^{73}Se 0.89%	^{74}Se 119.78 d	^{75}Se 9.37%	^{76}Se	
	^{62}As ?	^{63}As < 43 1e-09	^{64}As 40 ms	^{65}As 170 ms	^{66}As 95.77 ms	^{67}As 42.5 s	^{68}As 151.6 s	^{69}As 15.2 m	^{70}As 52.6 m	^{71}As 65.30 h	^{72}As 26.0 h	^{73}As 80.30 d	^{74}As 17.77 d	^{75}As 100%
32	^{61}Ge 44 ms	^{62}Ge 129 ms	^{63}Ge 142 ms	^{64}Ge 63.7 s	^{65}Ge 30.9 s	^{66}Ge 2.26 h	^{67}Ge 18.9 m	^{68}Ge 270.93 d	^{69}Ge 39.05 h	^{70}Ge 20.57%	^{71}Ge 11.43 d	^{72}Ge 27.45%	^{73}Ge 7.75%	^{74}Ge 36.50%
	^{60}Ga 70 ms	^{61}Ga 167 ms	^{62}Ga 116.121 ms	^{63}Ga 32.4 s	^{64}Ga 2.627 m	^{65}Ga 15.2 m	^{66}Ga 9.304 h	^{67}Ga 3.2617 d	^{68}Ga 67.845 m	^{69}Ga 60.108%	^{70}Ga 21.14 m	^{71}Ga 39.892%	^{72}Ga 14.025 h	^{73}Ga 4.86 h
30	^{59}Zn 182.0 ms	^{60}Zn 2.38 m	^{61}Zn 89.1 s	^{62}Zn 9.193 h	^{63}Zn 38.47 m	^{64}Zn 49.17%	^{65}Zn 243.93 d	^{66}Zn 27.73%	^{67}Zn 4.04%	^{68}Zn 18.45%	^{69}Zn 13.756 h, 56.4 m	^{70}Zn 0.61%	^{71}Zn 4.125 h, 2.45 m	^{72}Zn 46.5 h
	^{58}Cu 3.204 s	^{59}Cu 81.5 s	^{60}Cu 23.7 m	^{61}Cu 3.339 h	^{62}Cu 9.67 m	^{63}Cu 69.15%	^{64}Cu 12.7004 h	^{65}Cu 30.85%	^{66}Cu 5.120 m	^{67}Cu 61.83 h	^{68}Cu 3.75 h, 30.9 s	^{69}Cu 2.85 m	^{70}Cu 3.5 s, 44.5 s	^{71}Cu 19.4 s
28	^{57}Ni 35.60 h	^{58}Ni 68.077%	^{59}Ni 81 ky	^{60}Ni 26.223%	^{61}Ni 1.1399%	^{62}Ni 3.6346%	^{63}Ni 101.2 y	^{64}Ni 0.9255%	^{65}Ni 2.5175 h	^{66}Ni 54.6 h	^{67}Ni 21 s	^{68}Ni 29 s	^{69}Ni 3.5 s, 11.4 s	^{70}Ni 6.0 s
	^{56}Co 77.236 d	^{57}Co 271.70 d	^{58}Co 9.10 h, 70.86 d	^{59}Co 100%	^{60}Co 10.467 m, 5.2712 y	^{61}Co 1.649 h	^{62}Co 13.86 m, 1.54 m	^{63}Co 26.9 s	^{64}Co 300 ms	^{65}Co 1.16 s	^{66}Co 194 ms	^{67}Co 329 ms	^{68}Co 1.6 s, 200 ms	^{69}Co 180 ms
	30	32	34	36	38	40	42							

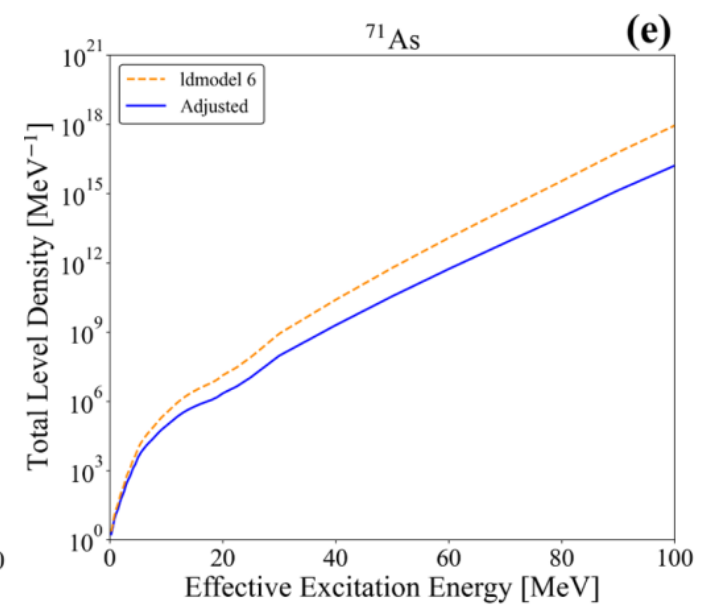
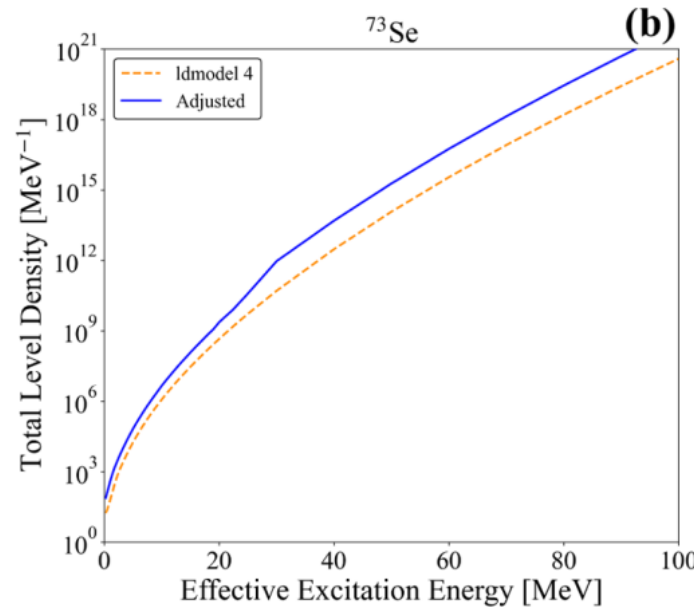
*M. B. Fox et al., PRC, 103(3):034601, 2021 & PRC 104, 064615 (2021) ²⁴

Level Density Adjustments in $^{75}\text{As}(p,x)$



$$\rho(E_x, J, \pi) = \exp(c\sqrt{E_x - \delta})\rho_{\text{mic}}(E_x - \delta, J, \pi) \quad \text{ctable}$$

34 73 0.24		
33 74 0.3		
33 73 0.75		
33 71 -0.4		
32 69 0.285		
31 67 -0.45		
<hr/>		
34 73 -0.65		
34 72 0.14		
33 73 -1.85		
32 69 -0.25		
31 67 5.5		

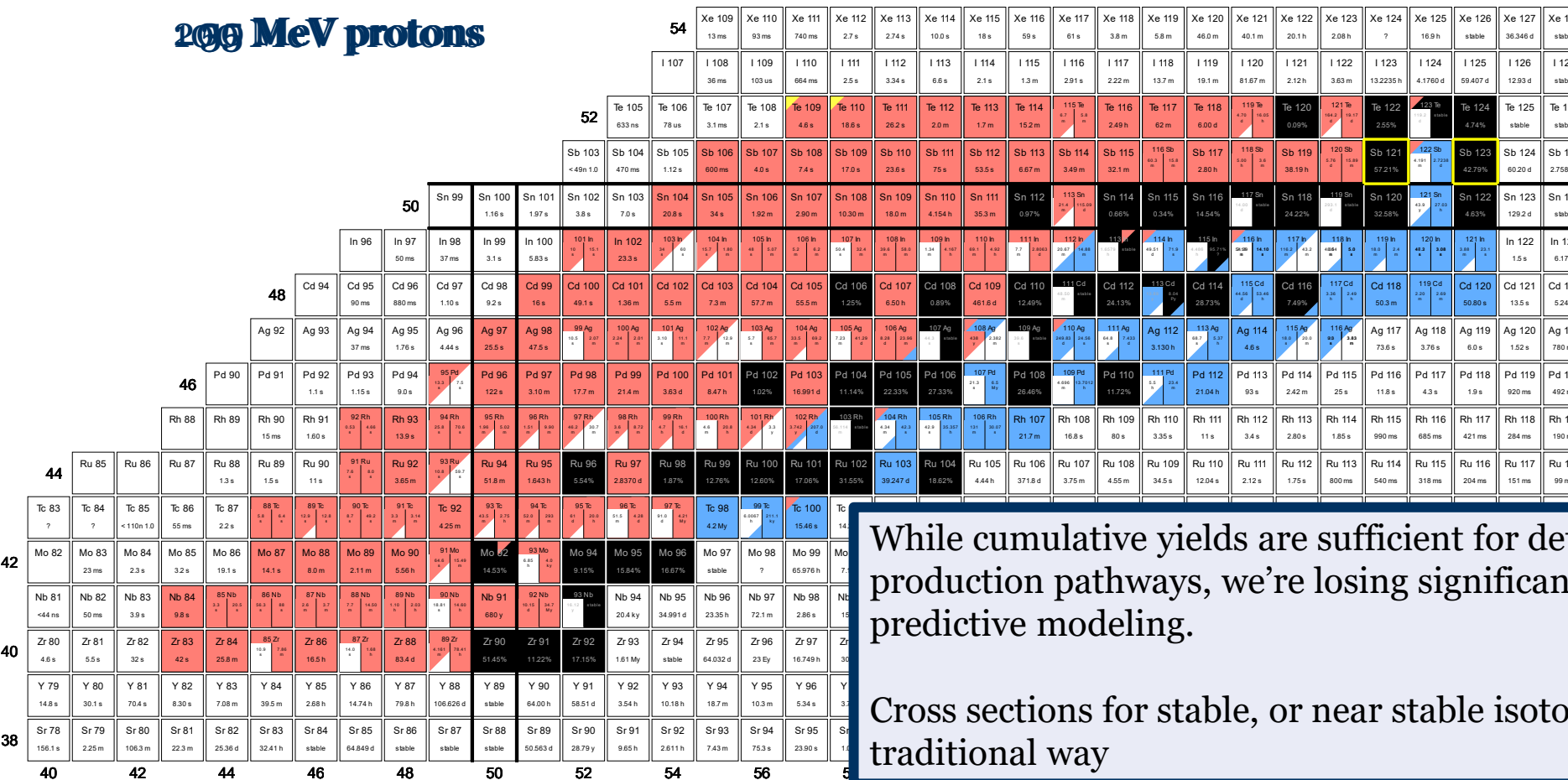


We see global χ^2 improvements of ~ 3 , but need more discrete level data and level density measurements!

Stacked-target experiments for measuring cross sections

As we increase in incident proton energy, from 35 MeV all the way to 200 MeV, we open channels for 200+ potential products.

200 MeV protons



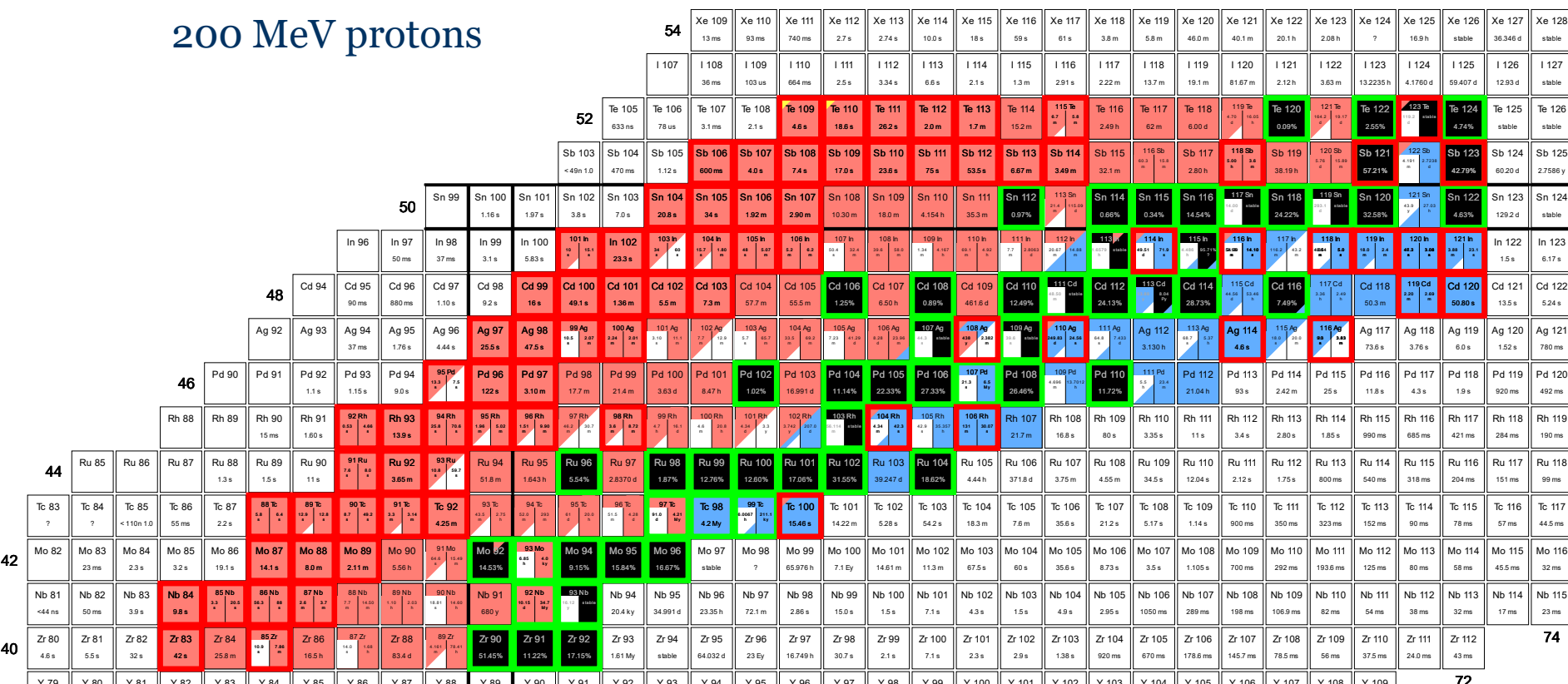
While cumulative yields are sufficient for determining medical isotope production pathways, we're losing significant granularity in terms of predictive modeling.

Cross sections for stable, or near stable isotopes cannot be measured in this traditional way

Stacked-target experiments for measuring cross sections

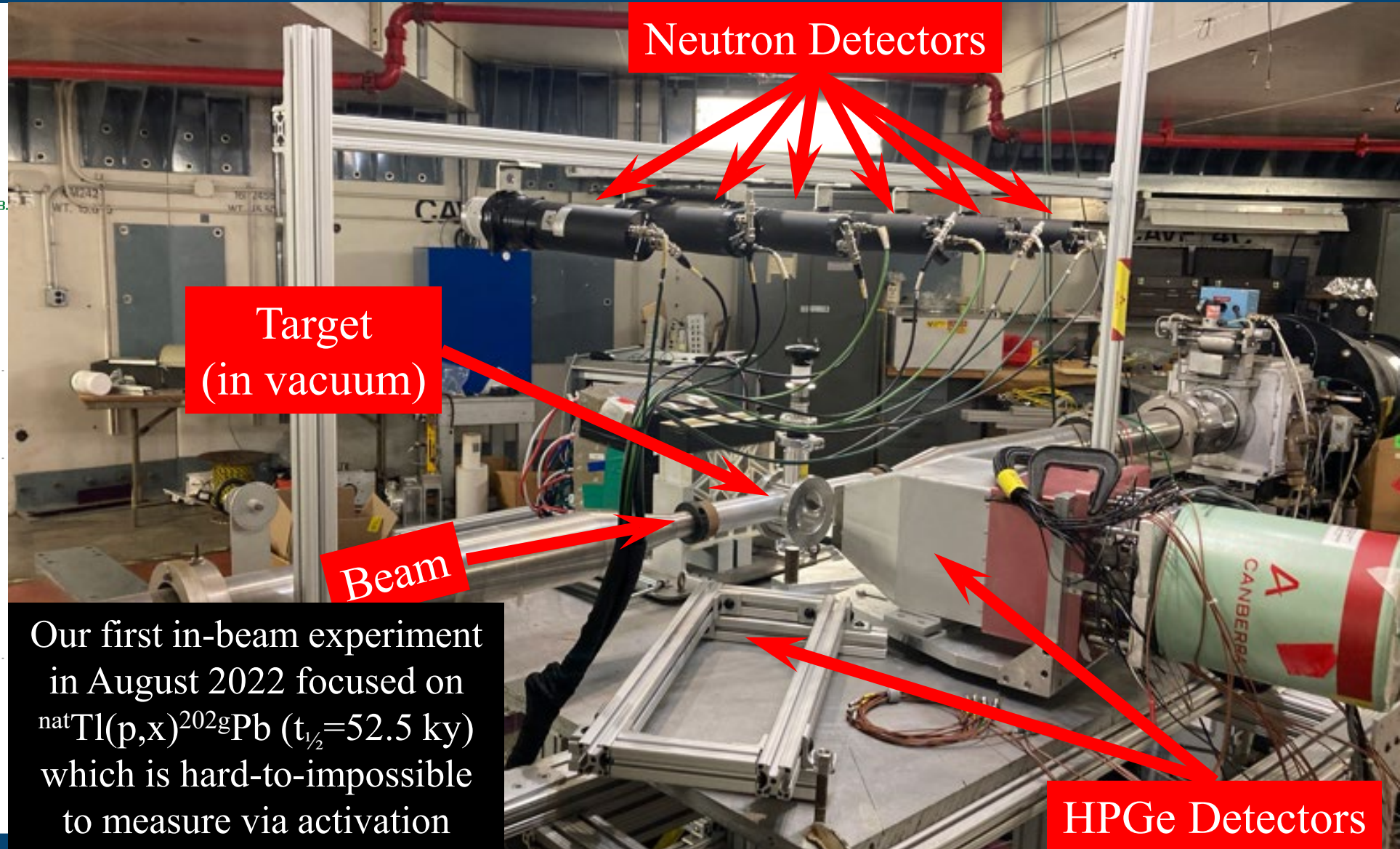
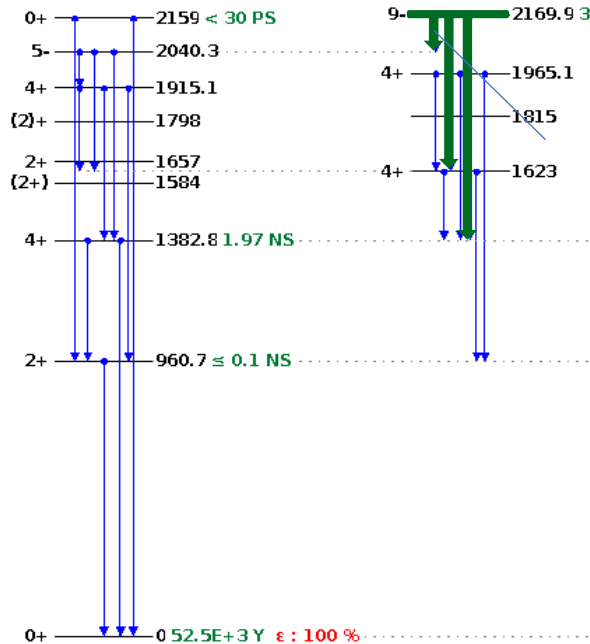
As we increase in incident proton energy, from 35 MeV all the way to 200 MeV, we open channels for 200+ potential products.

200 MeV protons



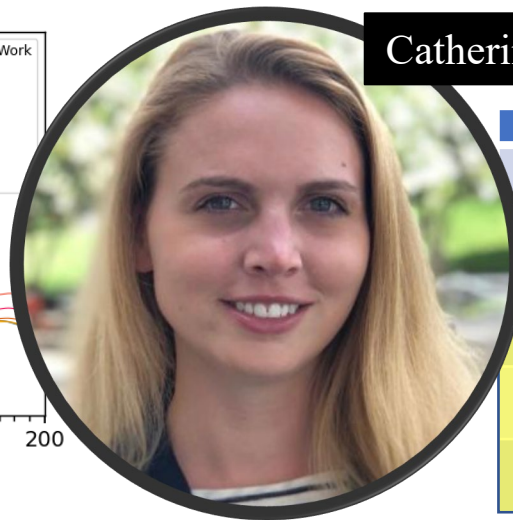
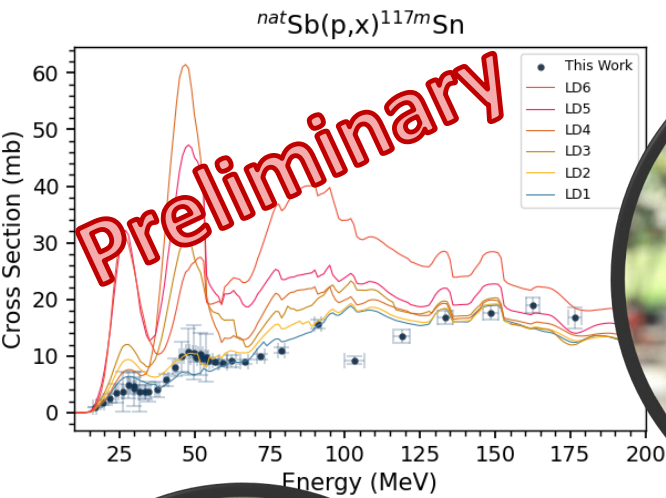
It's not possible to get independent cross section measurements for many of the products produced using the stacked target approach, so a different approach is needed!

Recent measurement of $Tl(p,xg)^{202}Pb$ via prompt gamma ray spectroscopy



Limitations to Residual Product-Based Fitting

- This approach can't be used to fit to any products which are stable, lack observable decay gammas, or are too short-lived (< 10ish minutes)
 - We need something beyond the stacked-foil technique!



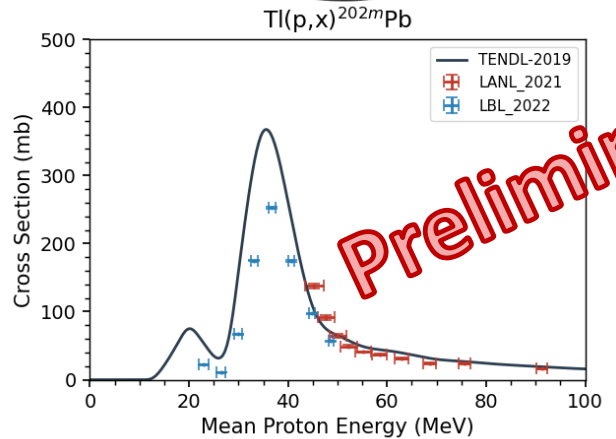
Catherine Apgar

Isotope Production

Isotope	Target	Beam	Measurement focus	Energy range	Site	Year
$^{72}\text{Se}, ^{68}\text{Ge}$	^{75}As	p	Primary: $^{72}\text{Se}, ^{68}\text{Ge}$ Impurities: $^{70,71,73,75}\text{Se}, ^{66,67,69,71}\text{Ge}$	Up to 200 MeV	All	1
^{202}Pb	^{205}Tl	p,d	Primary: $^{202m,202g}\text{Pb}$ Impurities: $^{198-205}\text{Pb}$	Up to 200 MeV	All	2
^{202}Pb	^{203}Tl	p,d	Primary: $^{202m,202g}\text{Pb}$ Impurities: $^{198-203}\text{Pb}$	Up to 200 MeV	All	3
$^{119}\text{Te}, ^{117m}\text{Sn}$	natSb	p,d	Primary: $^{119m,119g}\text{Te}, ^{117m}\text{Sn}$ Impurities: $^{116-118,121m/g,123m}\text{Te}, ^{113,119m,121m/g}\text{Sn}$	Up to 200 MeV	All	4
^{134}Ce	natLa	p,d	Primary: ^{134}Ce Impurities: $^{132,133,133,137,139}\text{Ce}$	Extend to 200 MeV	LANL, BNL	4



Michael Skulski



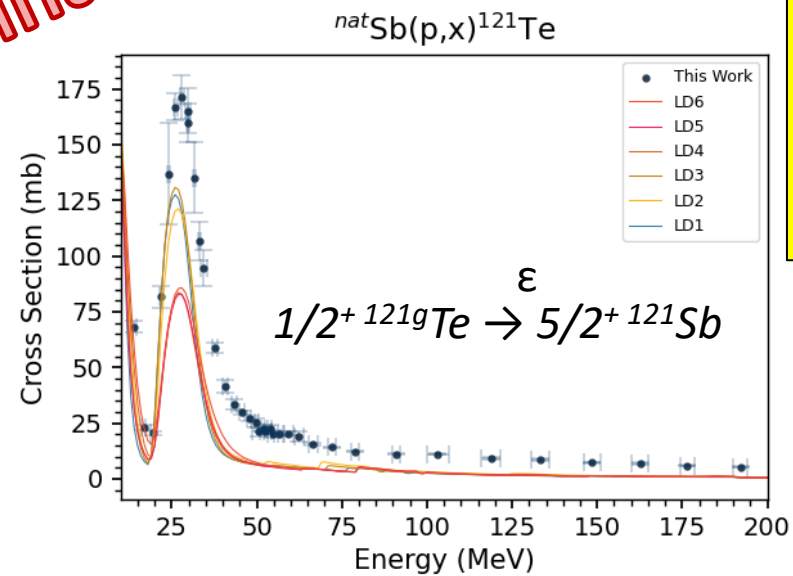
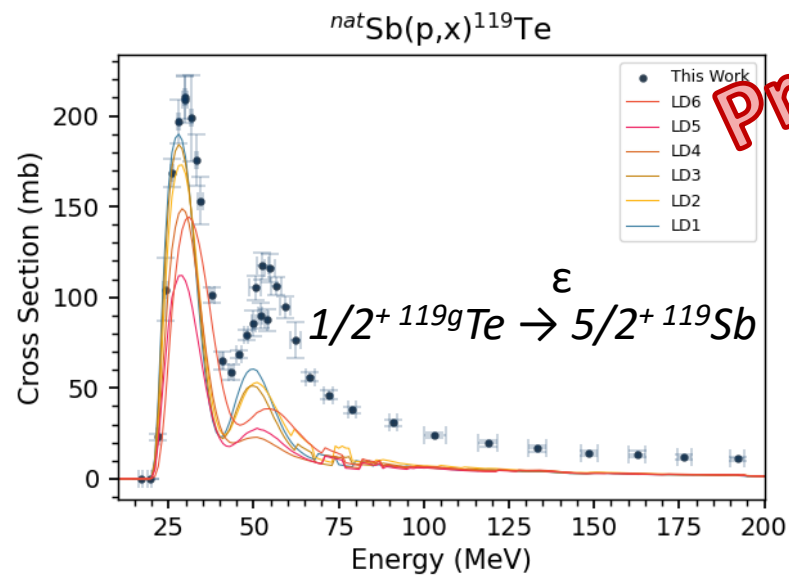
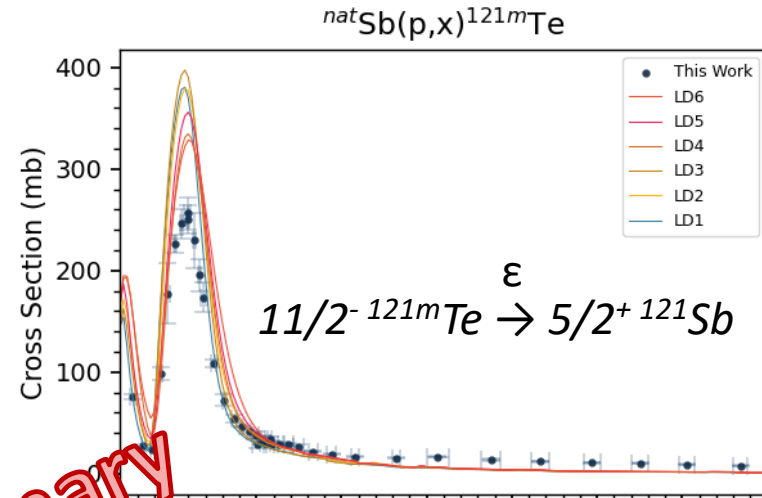
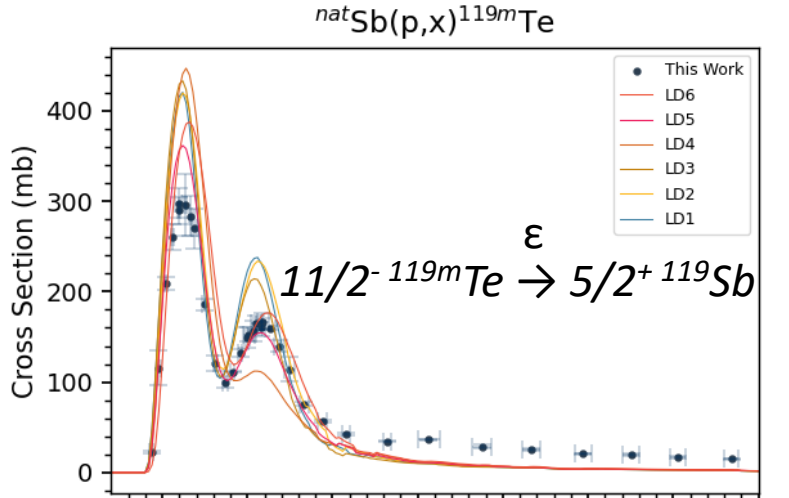
Stay tuned!

Flux Monitoring

Nuclear reaction	Energy range	Site	Year
$\text{natTi}(p,x)^{48}\text{V}$	Extend to 200 MeV	All	2
$\text{natNb}(p,4n)^{90}\text{Mo}$	Up to 200 MeV	BNL	Out years

*M. B. Fox *et al.*, *PRC*, 103(3):034601, 2021 & *PRC* 104, 064615 (2021) ²⁹

Isomer Population Modeling for Sb(p,xn)



Preliminary

Both phenomenological & microscopic LD models overpredict Te isomer population and underpredict g.s., even with adjustments



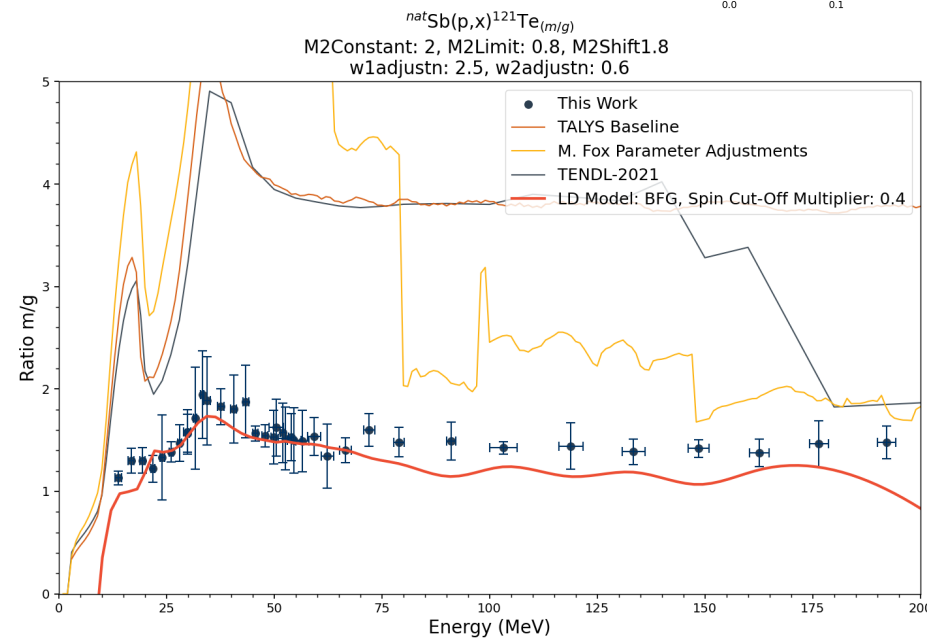
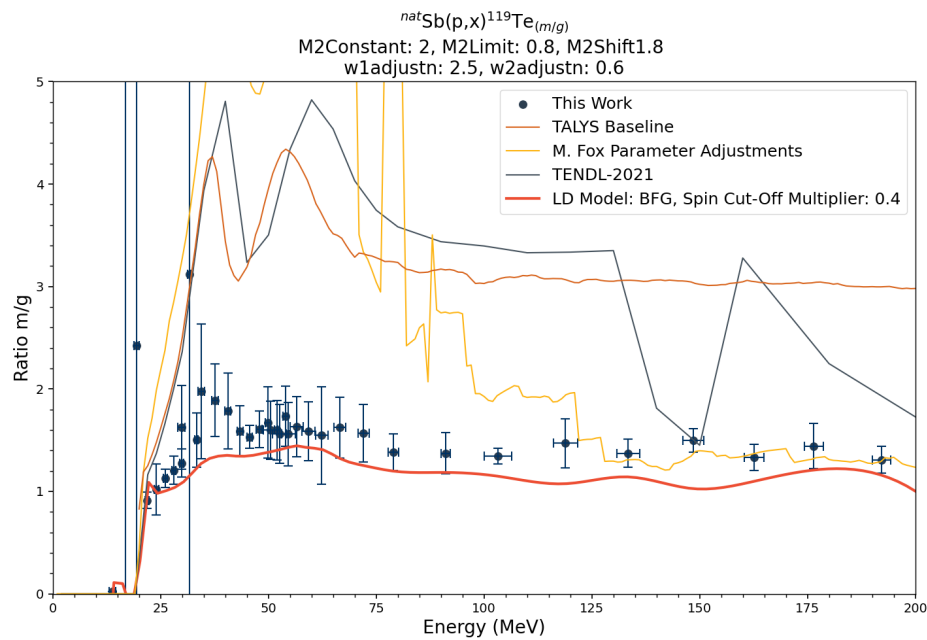
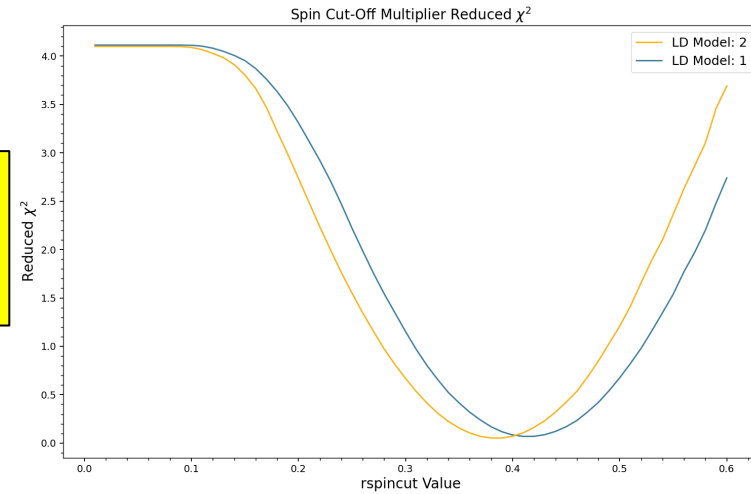
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Adjustments to the Spin Cut-Off Parameter (BFM / 1dmodel 2)

- TALYS provides the variables **Rspincut**, which functions as a global adjustment applied to the spin cut-off parameter:

$$\sigma_F^2(E_x) = 0.01389 \frac{A^{5/3}}{\tilde{a}} \sqrt{aU}$$

Local minimum near 0.4



Beyond Rspincut: Other Parameters to Consider

OMP adjustments to the imaginary volume term:

- **W1adjust, w2adjust** adjustments to imaginary volume term

PE reaction adjustments:

- **M2Constant, M2Limit, M2Shift** previously explored
- **Rpipi, rpinu, rnupi, rnuu** adjustments based on residual nucleon-nucleon interactions

$$W_V(E) = w_1^n \frac{(E - E_f^n)^2}{(E - E_f^n)^2 + (w_2^n)^2}$$

$$M_{\pi\pi}^2 = R_{\pi\pi} M^2,$$

$$M_{\nu\nu}^2 = R_{\nu\nu} M^2,$$

$$M_{\pi\nu}^2 = R_{\pi\nu} M^2,$$

$$M_{\nu\pi}^2 = R_{\nu\pi} M^2.$$

Variable	Default	Adjusted
$R_{\pi\nu}$	1	1.5
$R_{\pi\pi}$	1	1
$R_{\nu\pi}$	1	1.5
$R_{\nu\nu}$	1.5	1.5
W1adjust n	1	2.5
W2adjust n	1	0.6
M2Constant	1	2
M2Limit	1	0.8
M2Shift	1	1.8

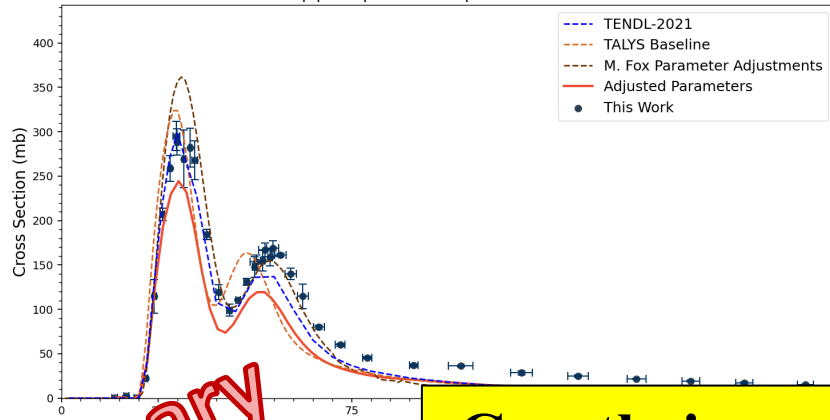
Preliminary



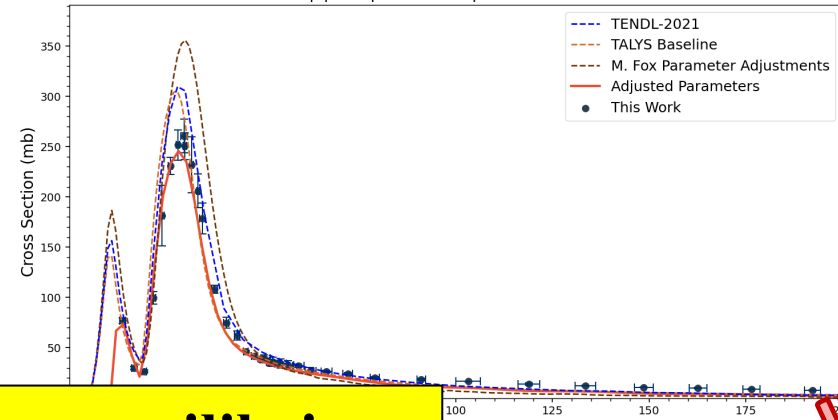
Catherine Apgar

Beyond Rspincut: Other Parameters to Consider

$^{nat}\text{Sb}(p,x)^{119m}\text{Te}$
 LD Model: 2, Spin Cut-Off Model: 2, Equidistant: y
 M2Constant: 2, M2Limit: 0.8, M2Shift1.8
 Spin Cut-Off Multiplier: 0.4 Colldamp: n preeqmode: 1 mpreeqmode:2 preeqspin: 1
 w1Adjust n: 2.5, w2Adjust n: 0.6
 rpipi: 1 rpinu: 1.5 rnu: 1.5 rnu: 1.5

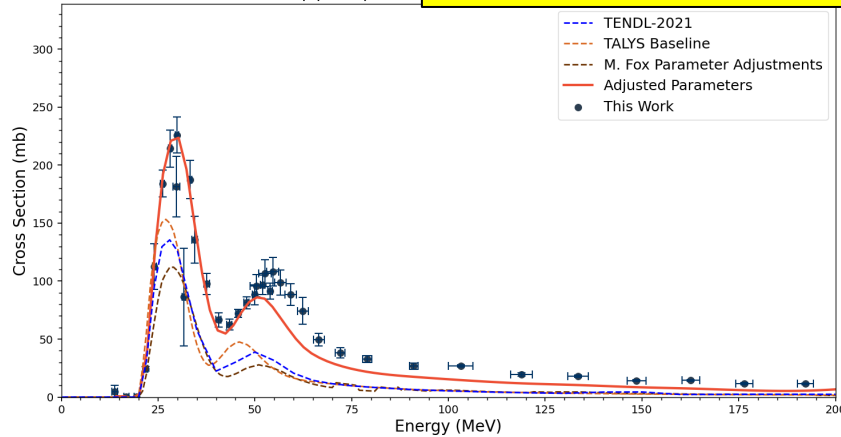


$^{nat}\text{Sb}(p,x)^{121m}\text{Te}$
 LD Model: 2, Spin Cut-Off Model: 2, Equidistant: y
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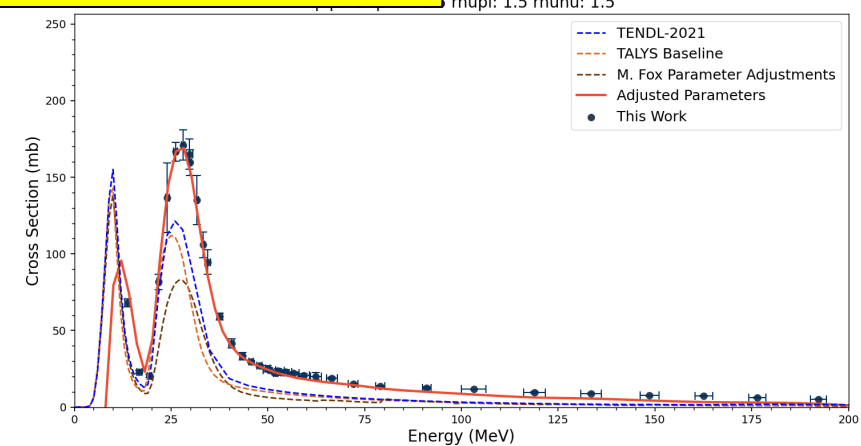


Greatly improved fitting of pre-equilibrium tail for higher incident energies!

$^{nat}\text{Sb}(p,x)^{119}\text{Te}$
 LD Model: 2, Spin Cut-Off Model: 2, Equidistant: y
 M2Constant: 2, M2Limit: 0.8, M2Shift1.8
 Spin Cut-Off Multiplier: 0.4 Colldamp: n preeqmode: 1 mpreeqmode:2 preeqspin: 1
 w1Adjust n: 2.5, w2Adjust n: 0.6
 rpipi: 1 rpinu: 1.5 rnu: 1.5 rnu: 1.5

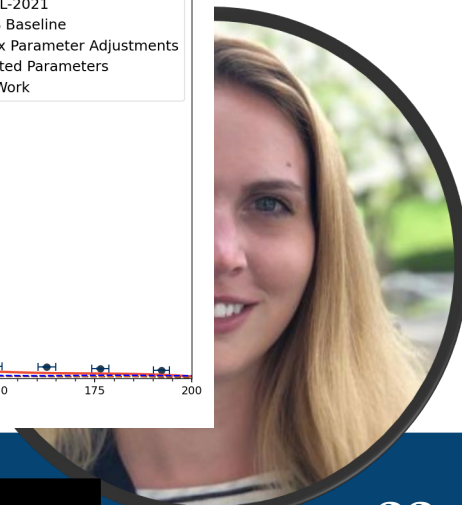


$^{nat}\text{Sb}(p,x)^{121}\text{Te}$
 LD Model: 2, Spin Cut-Off Model: 2, Equidistant: y
 M2Constant: 2, M2Limit: 0.8, M2Shift1.8
 Spin Cut-Off Multiplier: 0.4 Colldamp: n preeqmode: 1 mpreeqmode:2 preeqspin: 1
 w1Adjust n: 2.5, w2Adjust n: 0.6
 rpipi: 1 rpinu: 1.5 rnu: 1.5 rnu: 1.5



Preliminary

Preliminary



Thank you for your attention!

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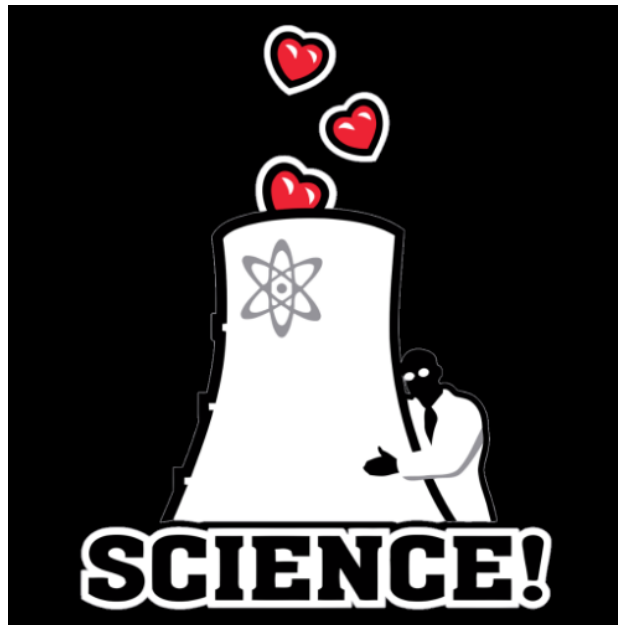
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