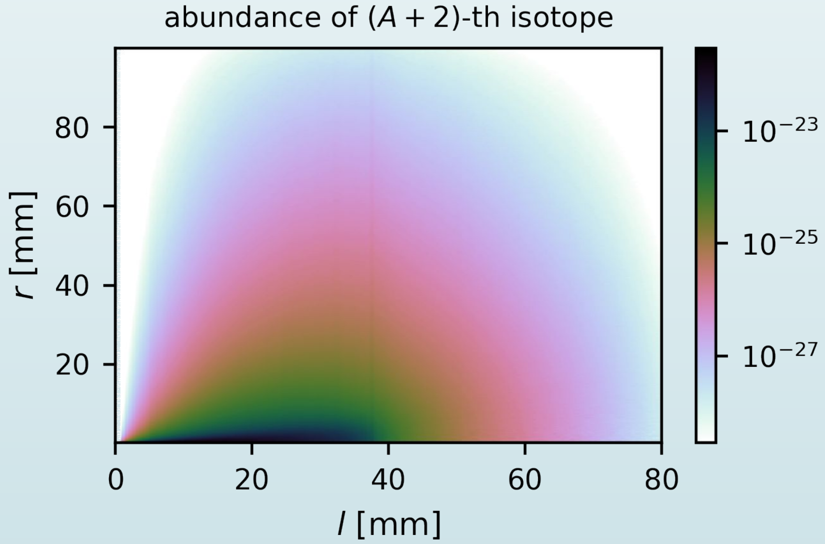
On feasibility of the laboratory *r*-process studies with laser-driven neutron source

Unfeasibility of the laboratory

Vojtěch Horný (ELI NP)

17th April 2024

Trieste (Italy)



My co-authors
Julien Fuchs, Sophia N. Chen,
Laurent Gremillet, Xavier Davoine

Joint ICTP-IAEA Workshop on Advanced Technologies in Laser-Driven Radiation Sources and Their Applications

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- What is *r*-process?
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- Laser-driven neutron source
  - Considered nuclear reactions
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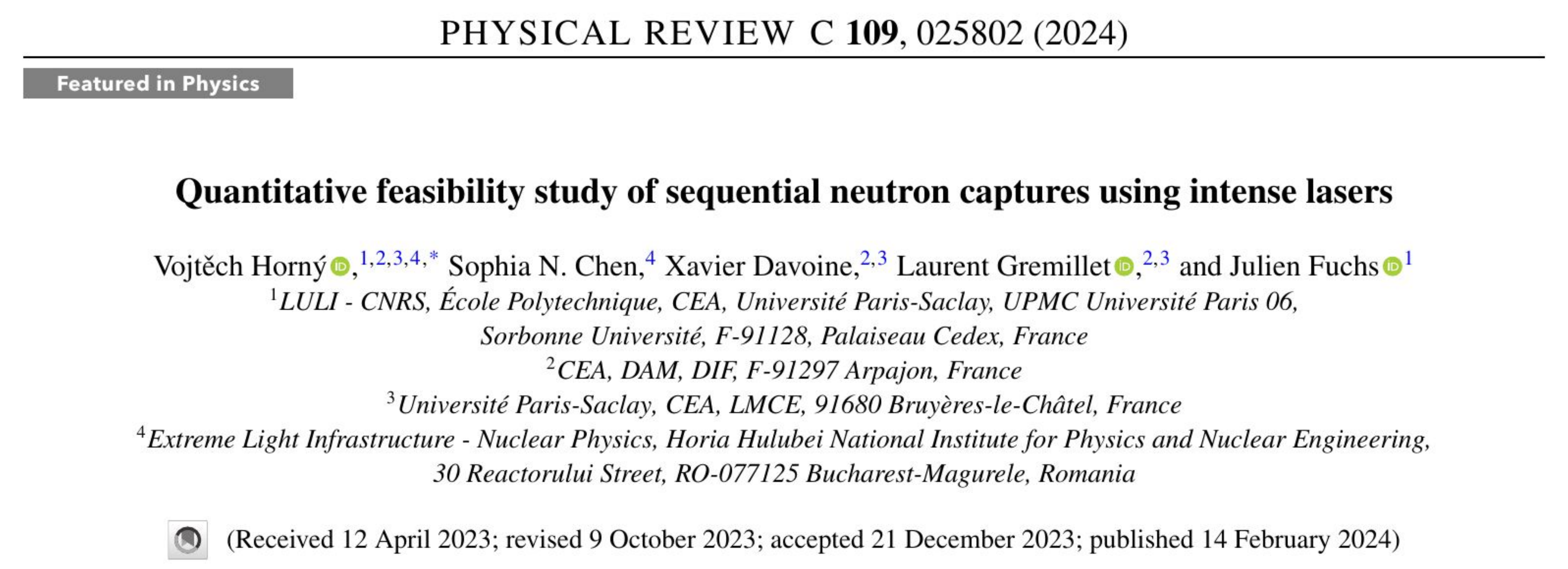
- Anticipated progress with a new generation of the high-power laser



PHYSICAL REVIEW C **109**, 025802 (2024)

Featured in Physics

Quantitative feasibility study of sequential neutron captures using intense lasers

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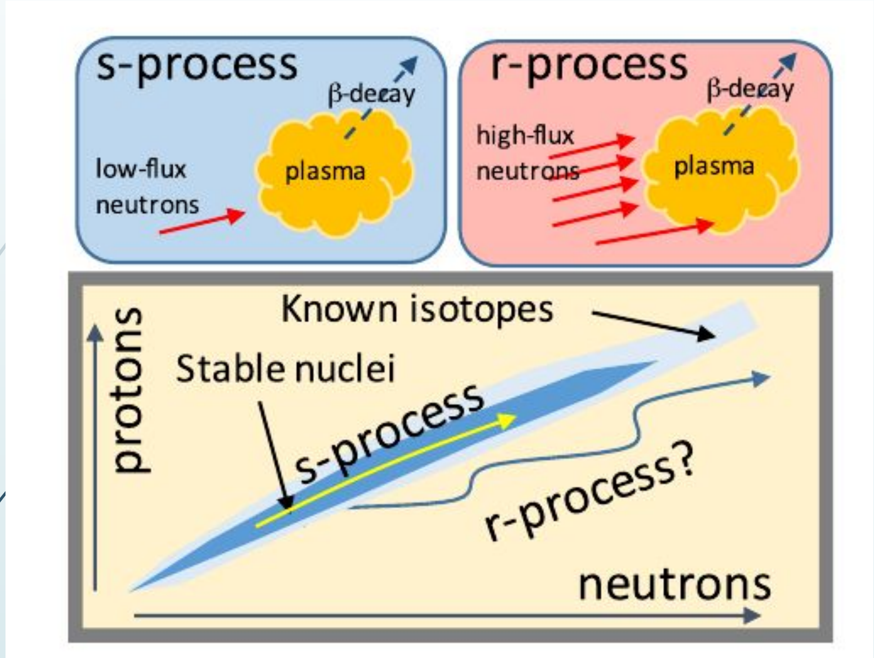
(Received 12 April 2023; revised 9 October 2023; accepted 21 December 2023; published 14 February 2024)

s-process and astrophysical *r*-process



s-process and β^--decay

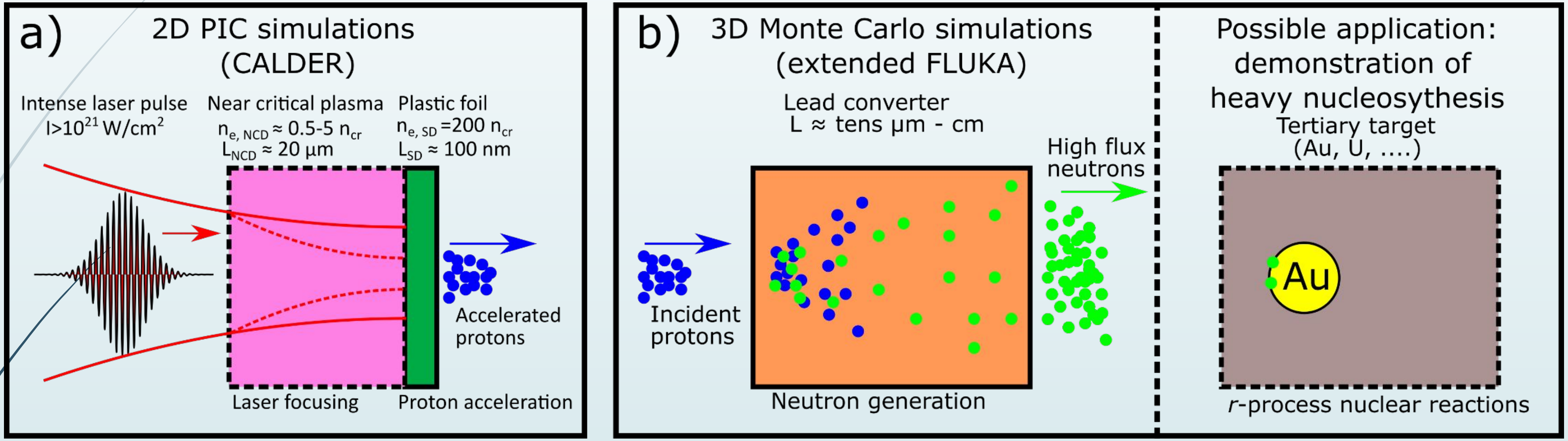
$$ {}^{A}_{Z}X + {}^{1}_{0}n \rightarrow {}^{A+1}{}_{Z}X \rightarrow {}^{A+1}_{Z+1}Y + e^- + \bar{\nu}_e $$

s-process works only up to ^{209}Bi, because ^{210}Po undergoes a-decay

$$ ^{210}\text{Po} \rightarrow {}^{206}\text{Pb} + {}^{4}\text{a} $$

- Rapid neutron capture: capture of two or more neutrons on the seed nucleus within the lifetime of the intermediate product.
- Origin of half of the atomic nuclei heavier than iron, and of the heavier isotopes of heavy elements.
- Occurs in *neutron star mergers*, probably not during *core-collapse of supernovae* as though till LIGO.
- Astrophysical conditions: extreme neutron density of >10^{20} cm^{-3}, temperature of ~keV.
- On Earth observed by the thermonuclear explosion.

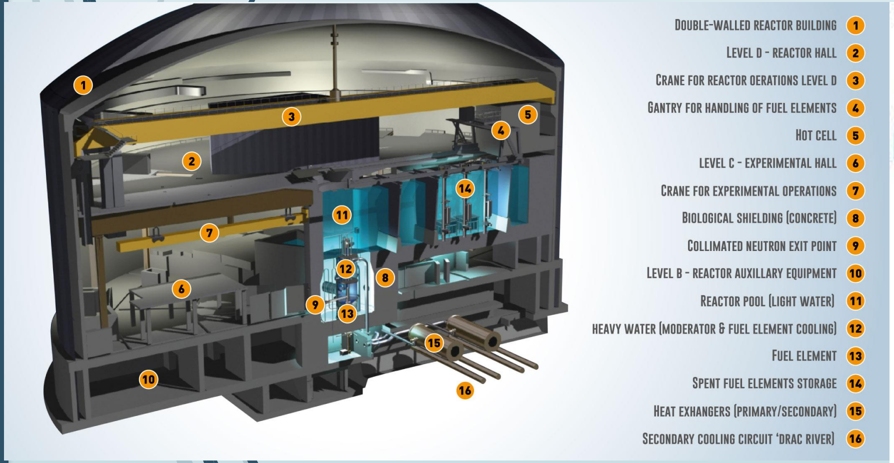
Idea of laboratory *r*-process study

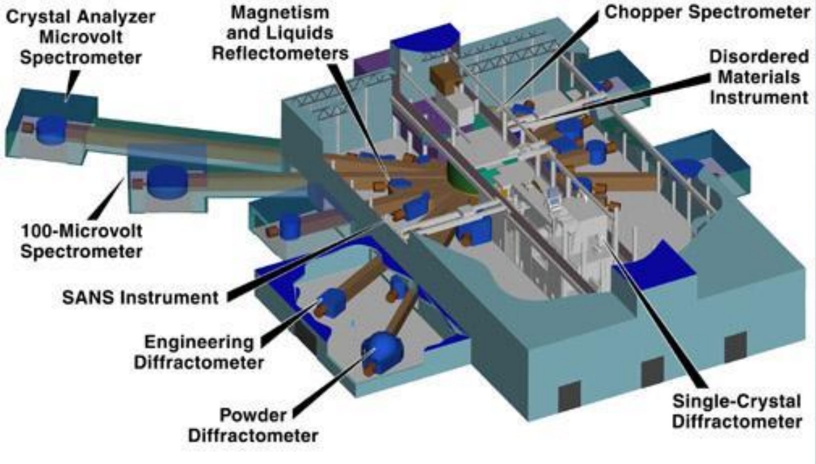


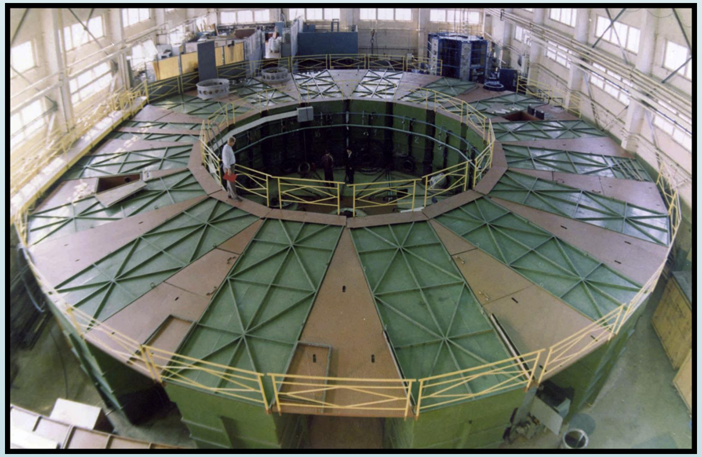
- Ion acceleration: TNSA, RPA, shock acceleration
- Conversion in the neutron converter: Be(d,n), Li(p,n) fusion reactions, spallation on heavy nuclei
- Neutron capture on an appropriate nucleus: e.g. ^{197}Au+2n -> ^{199}Au within the lifetime of intermediate product of ^{198}Au
- Measurement of decay of final product ^{199}Au

Contemporary neutron sources

Facility	Peak neutron flux [n cm^{-2} s^{-1}]	Average neutron flux [n cm^{-2} s^{-1}]	Neutron bunch duration	Repetition rate	Spectrum
Institut Laue–Langevin (reactor)	10^{15}	10^{15}	continuous	continuous	thermalised
Spallation Neutron source (accelerator)	10^{16}	10^{12}	1 μs	60 Hz	thermalised
National Ignition Facility (fusion devoted laser)	10^{26}	10^{10}	100 ps	once a day	MeVs
GIT-12 (Z-pinch)	10^{18}	10^{6}	10 ns	once a day	MeVs
Current lasers	10^{18}	10^{6}	1 ns	once per hour	broad



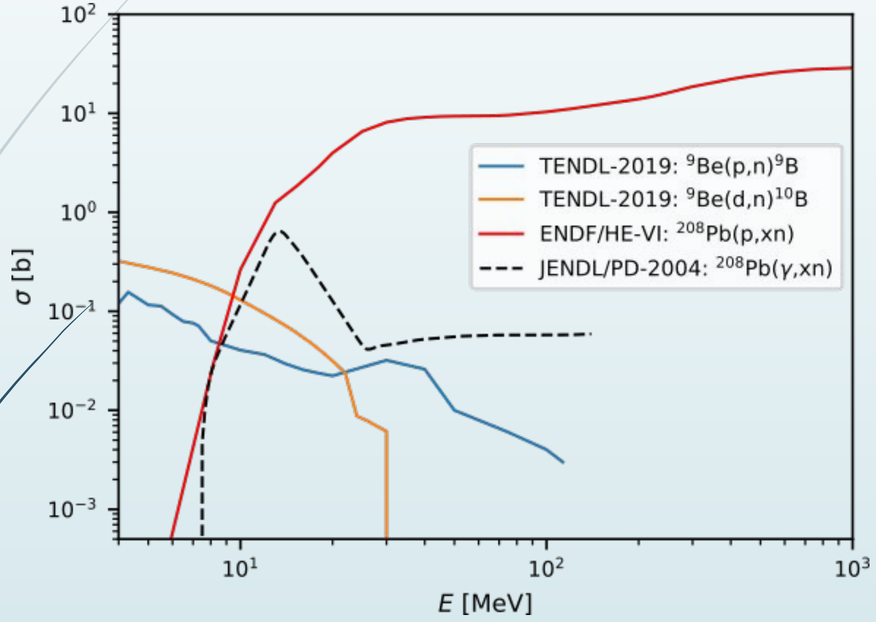




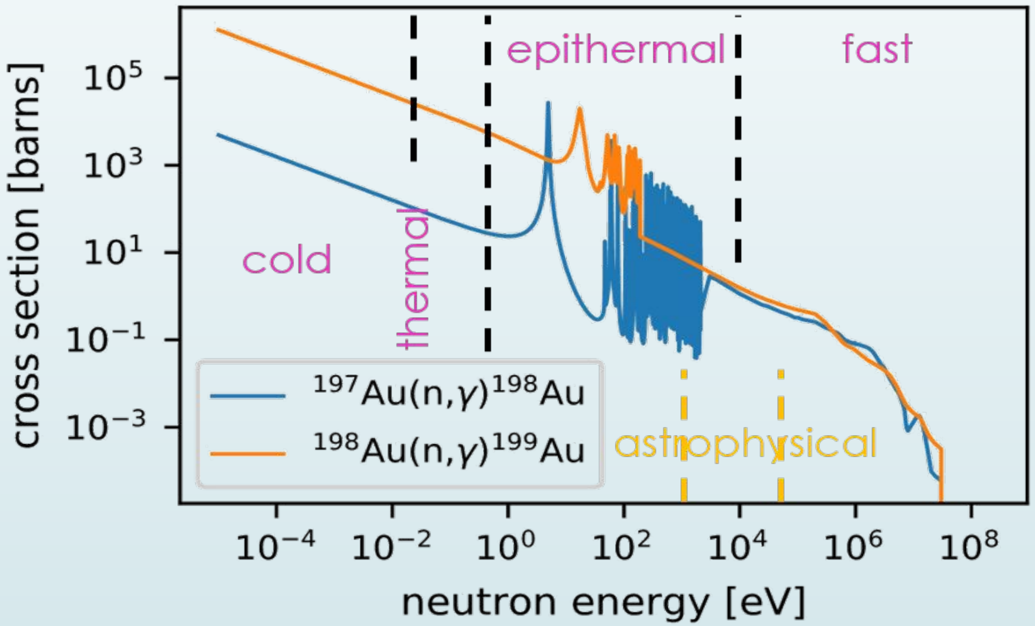
Cikhardt, J., et al. *Physics of Plasmas* 27.7 (2020): 072705.

Neutron generation and capture cross-sections

Conversion from ions to neutrons



Neutron capture



Source: TENDL-2019 database

Generation of neutrons

- Fusion reaction on light nuclei (keV-MeV).
- Spallation on heavy nuclei (MeVs)
- Photonuclear reactions from bremsstrahlung or synchrotron radiation (keV-MeV).
- Nuclear fission (MeV)

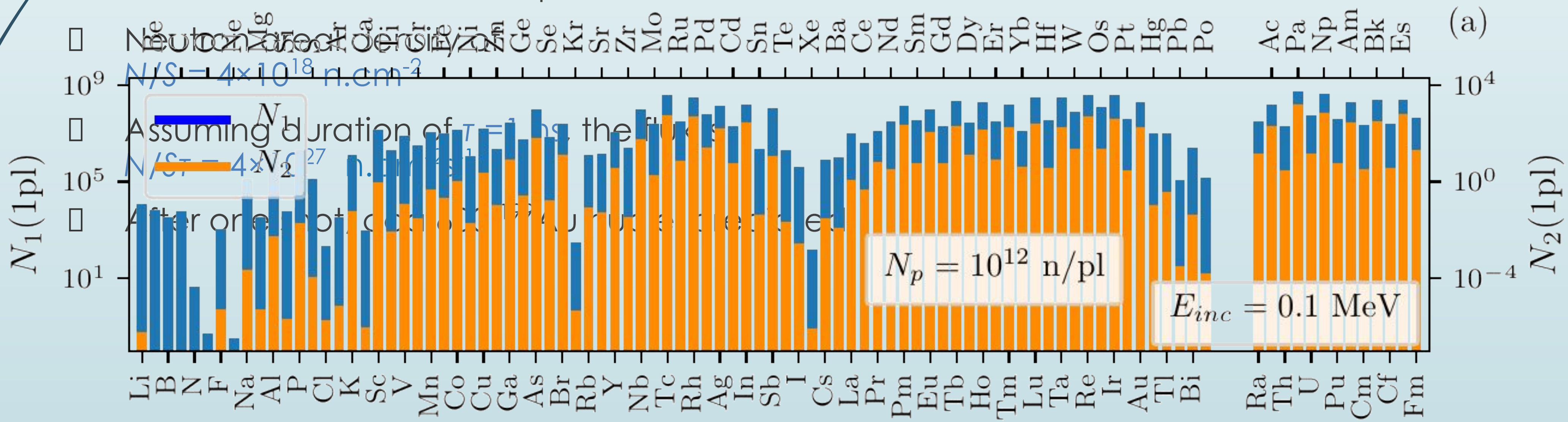
Uncertainities

- Lacking data especially A+1 isotopes → TALYS modelling.
- In astrophysics, increased cross-section
  - stellar enhancement factor

r-process with LDNS: recent literature

P. Hill & Y. Wu, PRC, 103, 014602 (2021)

- $N = 10^{12}$ neutrons per pulse
- Gaussian energy spectrum
  - 100 keV – 5 MeV
  - 10% relative width
- Uniform flux through $S = 25$ µm^2
- Converter thickness L=100 µm
- Neutron areal density of $N/S = 4\times10^{18}$ n.cm^{-2}
- Assuming duration of $\tau = 1$ ns, the flux is $N/S\tau = 4\times10^{27}$ n.cm^{-2}.s^{-1}
- After one shot, [illegible] Au nuclei [illegible] created



r-process with LDNS: recent literature

S. N. Chen et al., MRE, 4.5, 054402, 2019.

Neutron capture on ^{96}Zr

- Neutrons from LDNS
- In their estimation, it is assumed constant σ=1b (corresponds to ≈ 0.3 keV, for fast neutrons order of magnitude lower)
- Abundance of respective neutron-rich isotopes calculated.
- Taking similar parameters as previously
  - L = 100 μm
  - S = 25 μm^2
  - N = 10^{12} neutrons per pulse

  10^{-4} ^{98}Zr isotopes per shot is predicted.
- 10^5 neutron-rich isotopes in needed for measurement.

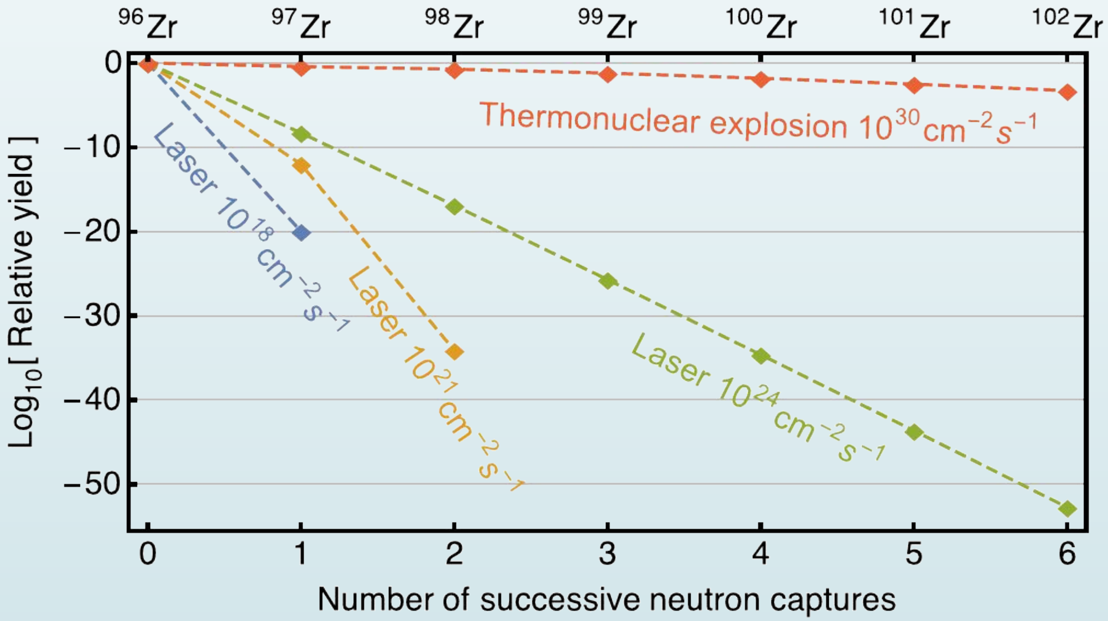
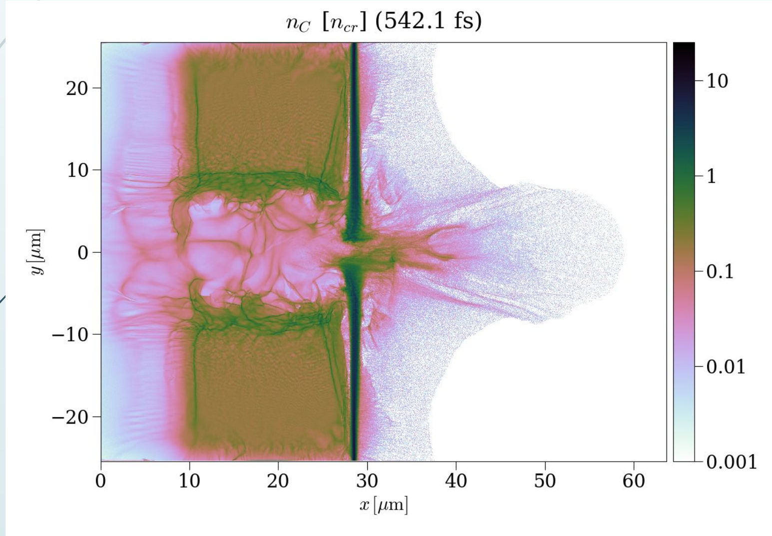


FIG. 8. Relative yields of isotopes produced by successive multi-neutron captures, i.e., by the r-process. The seed target is ^{96}Zr. The peak neutron flux is indicated for each curve. Laser experiments are calculated for a pulse duration of 1 ns. Also shown are the relative yields produced by a 1 μs long thermonuclear explosion.

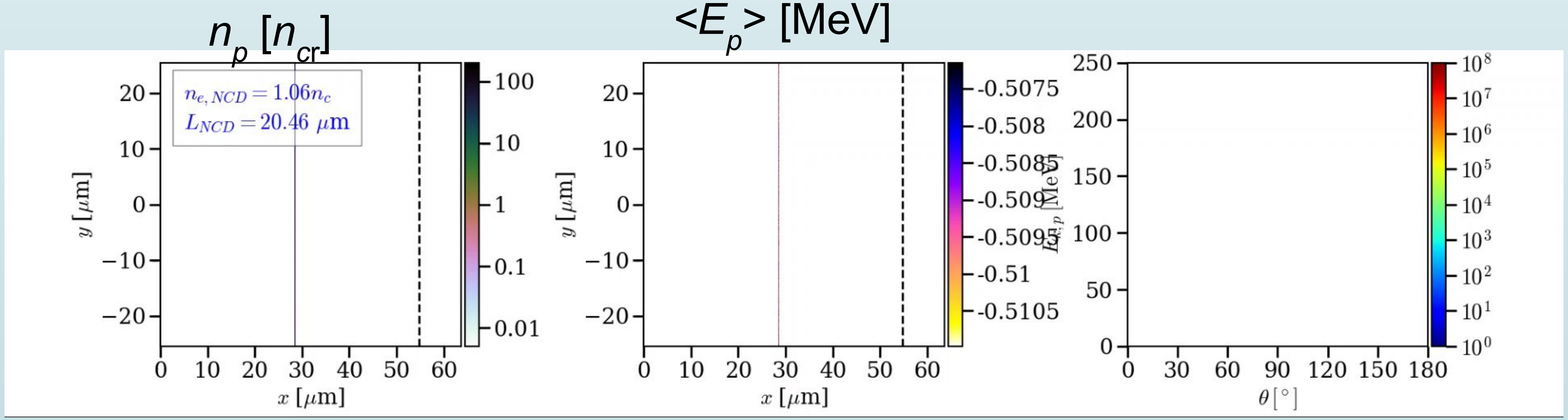
Realistic proton source: an optimized double-layer target for 1 PW Apollon



Horný, V., Chen, S.N., Davoine, X., Lelasseux, V., Gremillet, L. & Fuchs, J. (2022). High-flux neutron generation by laser-accelerated ions from single- and double-layer targets. *Scientific Reports, 12*, 19767

- Plasma: $n_{e,SD}$ = 200 n_{cr}, $n_{e,NCD}$ = 1.06 n_{cr}
  L_{SD} = 25 µm, L_{NCD} = 115 nm
- Laser: D_0 = 5 µm, a_0 = 30.6, τ = 20 fs

Tight laser self-focusing in near-critical plasma significantly enhances the proton acceleration.

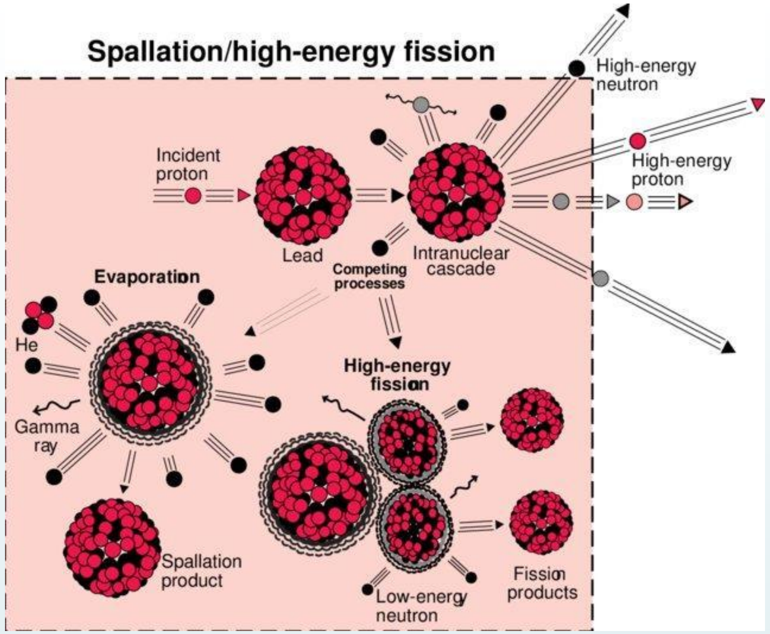


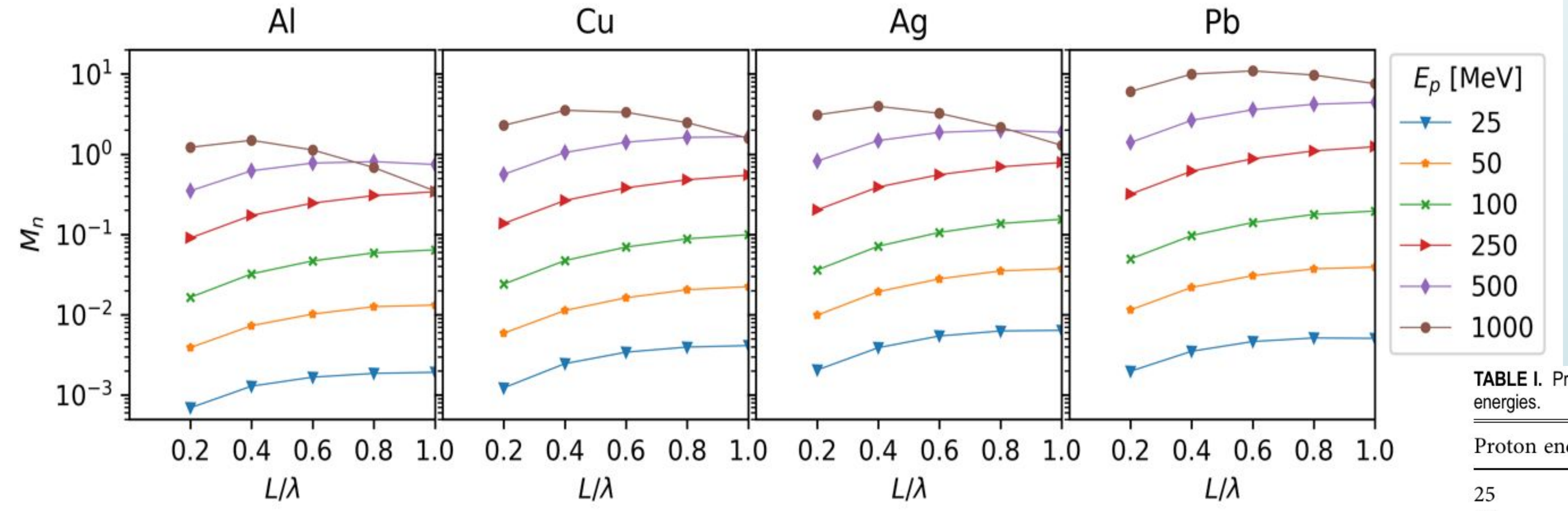
n_p [n_{cr}]

<E_p> [MeV]

Spallation

- A nuclear reaction in which the high-energy level of incident particles causes the nucleus to eject more than three particles, thus changing both its mass number and its atomic number.





cylinder
r = 50 cm

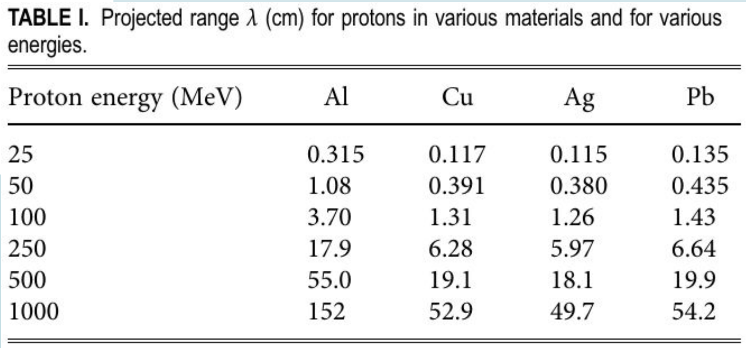


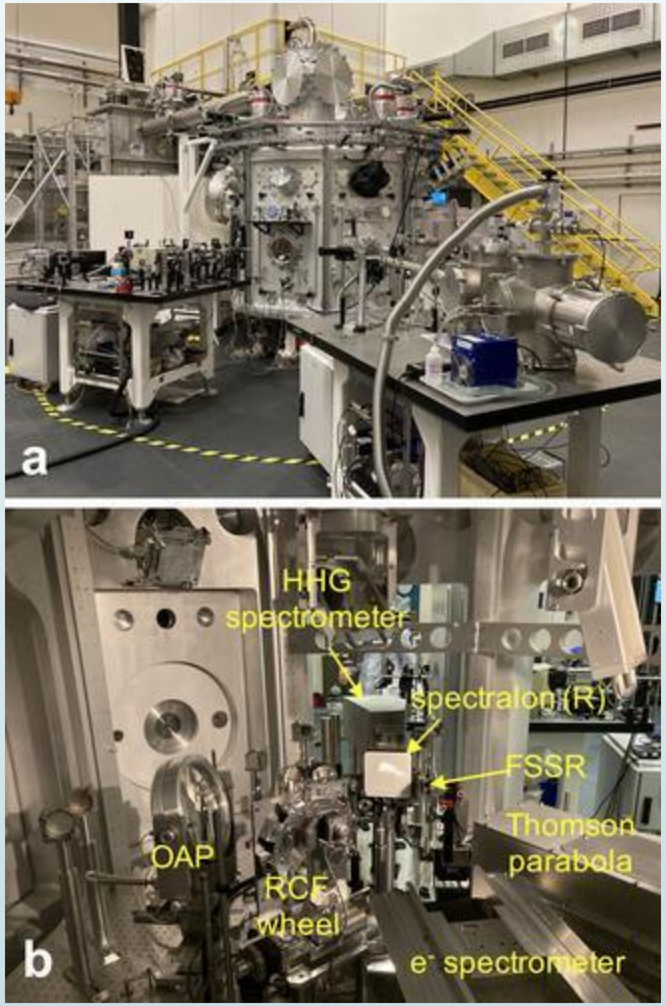
TABLE I. Projected range λ (cm) for protons in various materials and for various energies.

Proton energy (MeV)	Al	Cu	Ag	Pb
25	0.315	0.117	0.115	0.135
50	1.08	0.391	0.380	0.435
100	3.70	1.31	1.26	1.43
250	17.9	6.28	5.97	6.64
500	55.0	19.1	18.1	19.9
1000	152	52.9	49.7	54.2

Martinez, B., et al. Numerical investigation of spallation neutrons generated from petawatt-scale laser-driven proton beams. *Matter and Radiation at Extremes*, 2022, 7.2: 024401.
Kelley, K. C., Gadolinium-148 and Other Spallation Production CrossSection Measurements for Accelerator Target Facilities, Dissertation, Georgia Institute of Technology, 2004.

Expectation on the enhanced ion acceleration by Apollon or ELIs

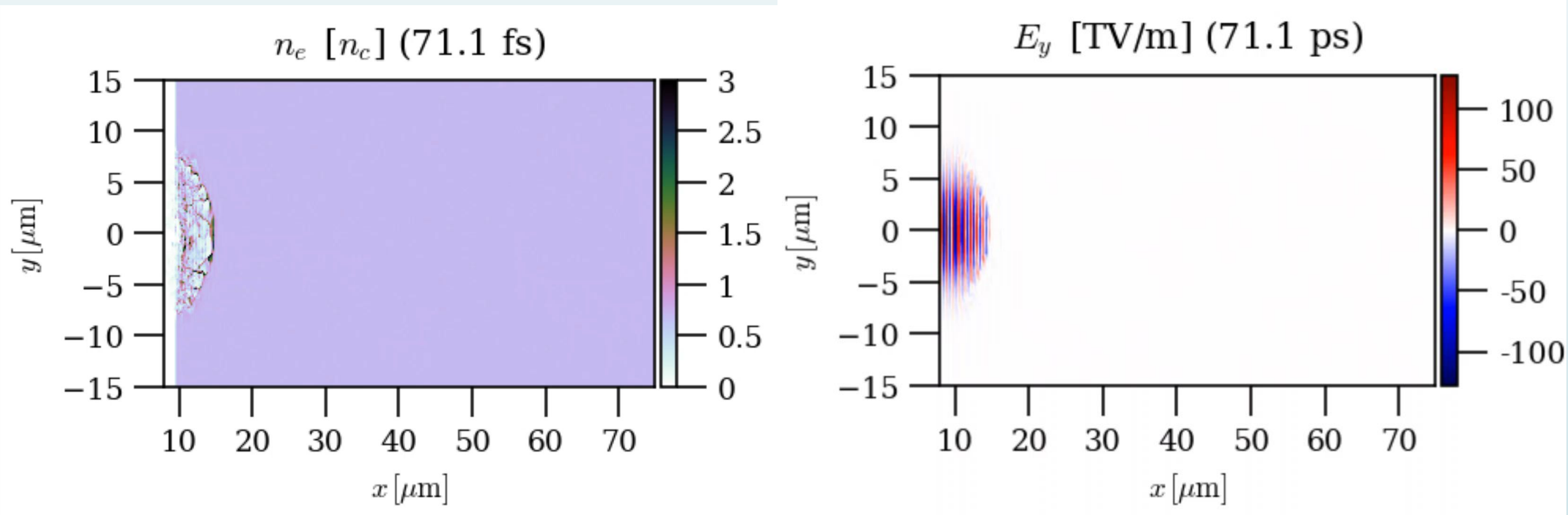
- We aim at the highest possible proton energies
  - **Relativistic transparency regime**
  - Mitigation of pre-pulses by **double plasma mirrors**.
  - **N**ear **C**ritical **D**ensity preplasma layer to intensify the laser pulse
- Apollon is already operational at 1 PW level.
- ELI NP operates at 1 PW and 10 PW.



Apollon's Short Focal Area
Burdonov, K., et al. "Characterization and performance of the Apollon Short-Focal-Area facility following its commissioning at 1 PW level." *Matter and Radiation at Extremes* 6.6 (2021): 064402.

Light intensification in NCD layer

- 2D PIC CALDER simulation of laser propagation in an uniform plasma with n_e=0.74 n_{cr}.
- Initial maximum intensity is 2×10^{21} W/cm^2.



- After 25.2 µm of propagation in the NCD plasma, the laser reaches a maximum intensity of 7.4×10^{21} W/cm^2, within a spot size of D = 1.33 µm.

Light intensification in NCD layer

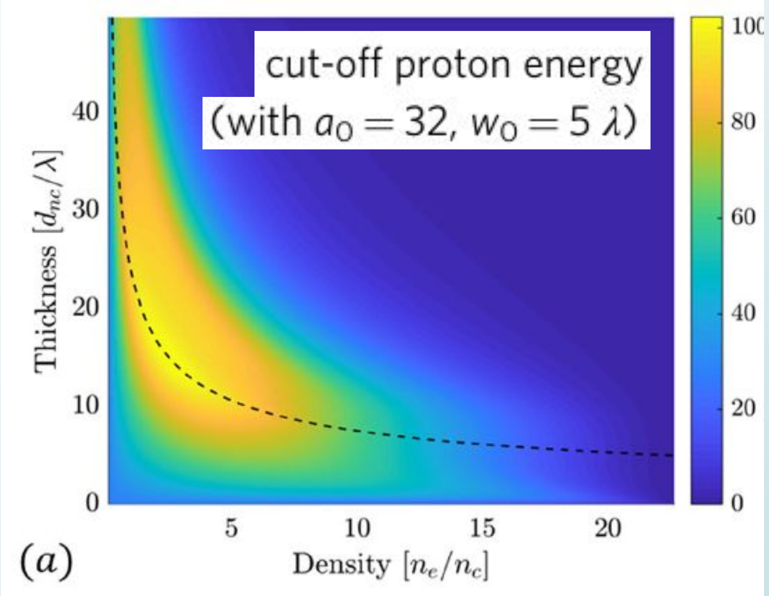
$$L_{\mathrm{NCD}} = 0.88\,\frac{D_0^2/\lambda_0}{(\tau_0 c/\lambda_0)^{1/3}}\,,$$

$$n_{e,\mathrm{NCD}} = 0.91\,\gamma_0 n_{\mathrm{cr}}\,\frac{\lambda_0^2}{D_0^2}\,(\tau_0 c/\lambda_0)^{2/3}$$

$$\gamma_0 = \sqrt{1 + a_0^2/2}$$

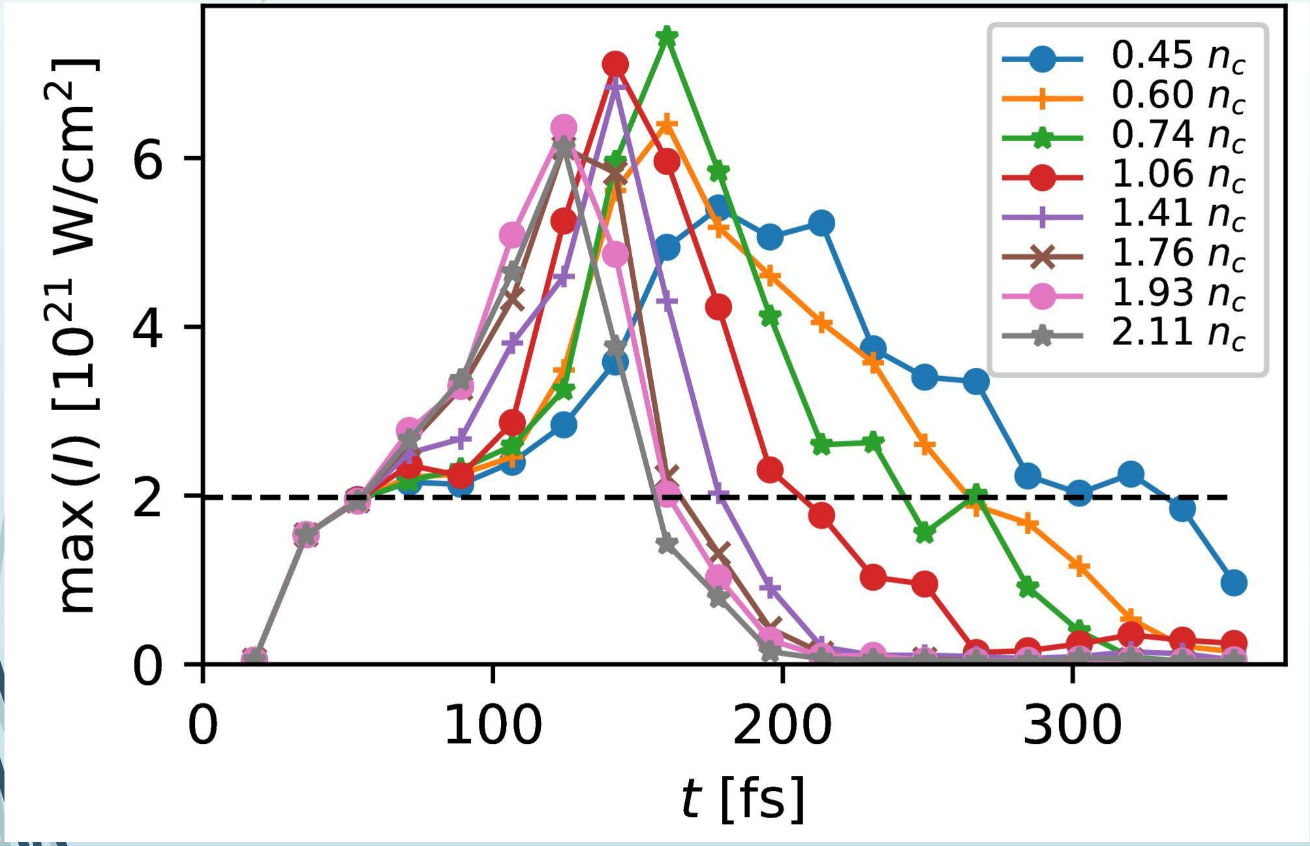
$$n_{\mathrm{cr}}\ [\mathrm{cm}^{-3}] = 1.1 \times 10^{21}/\lambda_0^2\ [\mu\mathrm{m}]$$

- Thin lens approximation predicts optimal focusing for Apollon 1 PW laser parameters:
  - n_e=1.93 n_{cr}
  - D_0=0.73 µm
  - L = 14.1 µm



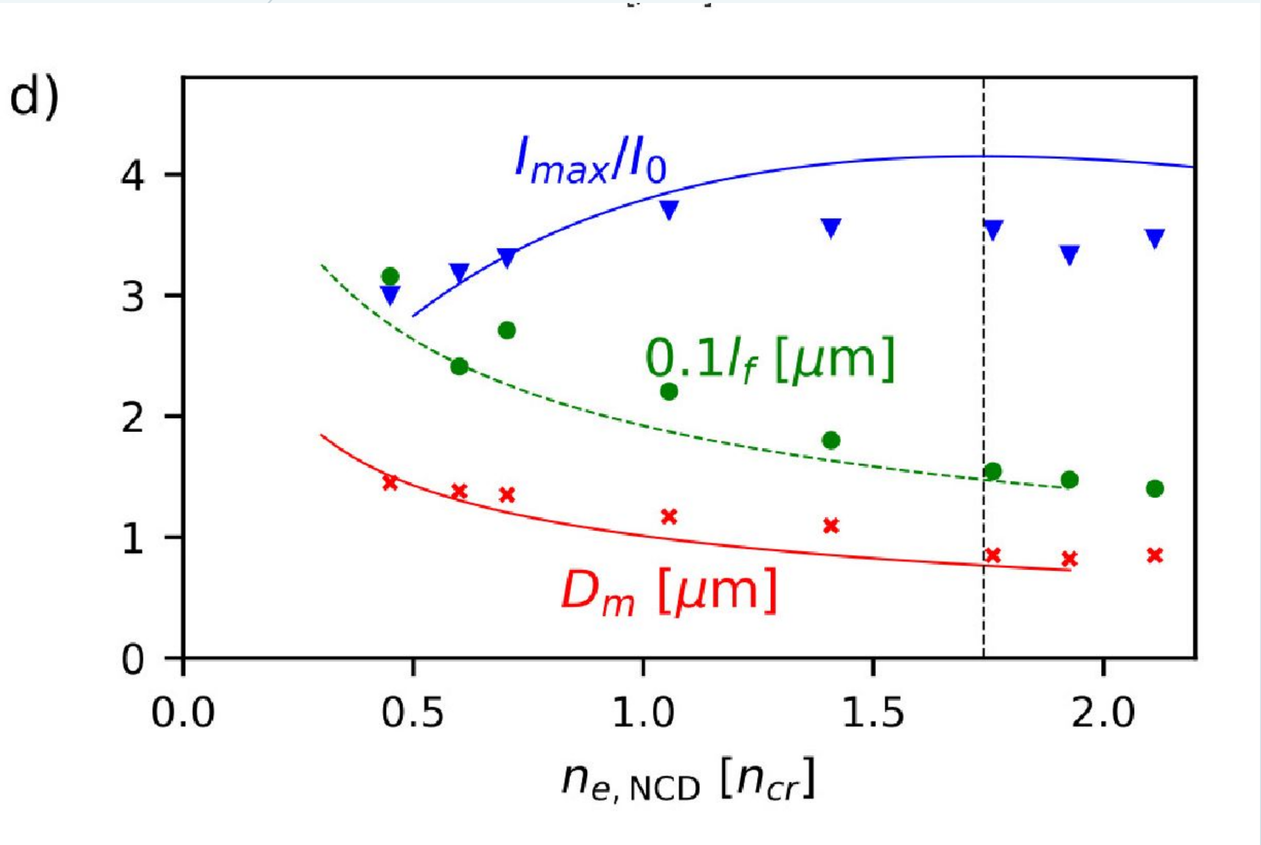
Pazzaglia, A., Fedeli, L., Formenti, A., Maffini, A., & Passoni, M. (2020). A theoretical model of laser-driven ion acceleration from near-critical double-layer targets. *Communications Physics*, 3(1), 1-13.

Light intensification in NCD layer



- Maximum intensification is around factor of three for NCD densities in range (0.60,2.11) n_{cr}.
- Optimum achieved for lower density of 0.74 n_{cr}.

Light intensification in NCD layer

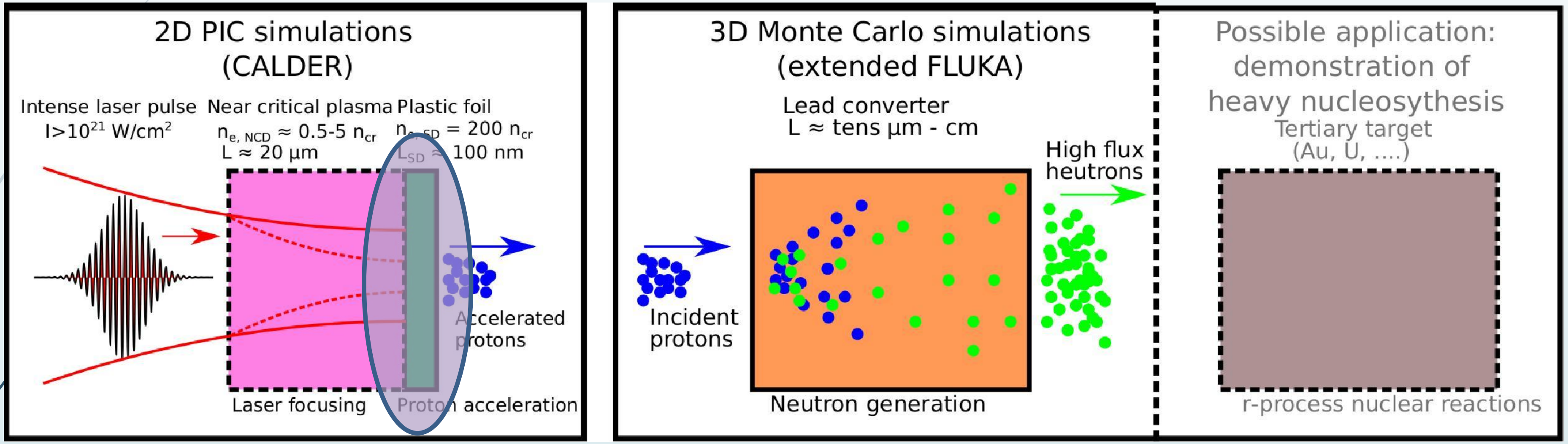


- Maximum intensification is around factor of three for NCD densities in range (0.60,2.11) n_{cr}.
- Optimum achieved for lower density of 0.74 n_{cr}.
- Best focusing agrees with model.

- The smallest laser spot does not necessarily lead to the highest intensity.

Pazzaglia, A., Fedeli, L., Formenti, A., Maffini, A., & Passoni, M. (2020). A theoretical model of laser-driven ion acceleration from near-critical double-layer targets. *Communications Physics*, *3*(1), 1-13.

Ultraintense-laser-based neutron generation

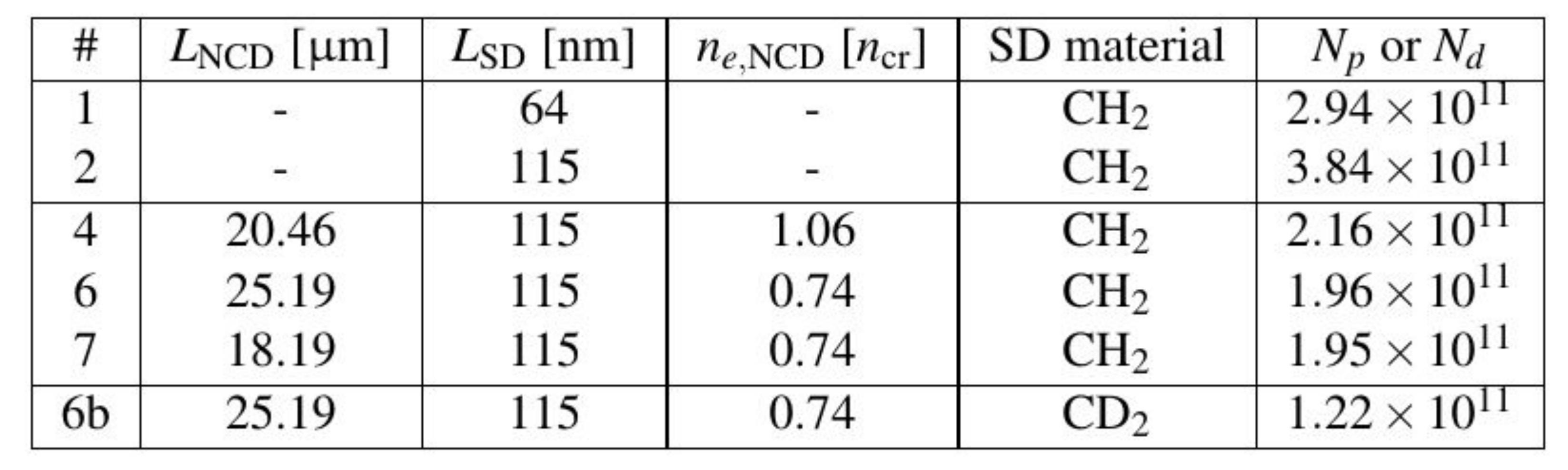


- High-energy protons from single-/double-layer targets driven by 1 PW Apollon laser
  - $E = 22$ J
  - $a_{0,d} = 30.6$
  - $\lambda_0 = 0.8$ µm
  - $D_0 = 5$ µm (FWHM of intensity)
  - $\tau = 20$ fs (FWHM of intensity)

1. nm-thick solid CH foil target, n_e=200 n_{cr}
2. µm-thick near-critical plasma layer + nm-thick solid CH foil target, n_e=200 n_{cr}

- Apollon or ELI NP systems operate once per minute - it is rather a nuclear security concern.
- In fact, similar systems could be capable of running at 10 Hz.

PIC simulations of proton acceleration



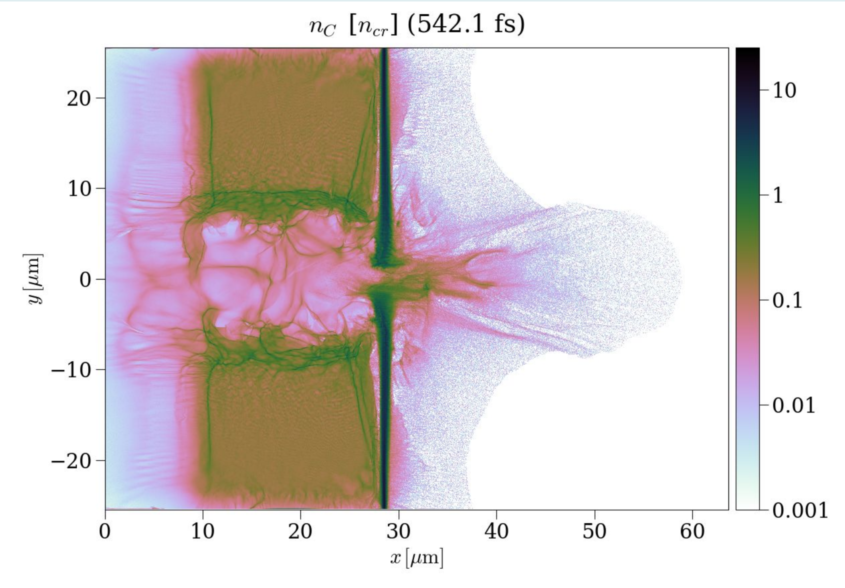
#	L_{NCD} [µm]	L_{SD} [nm]	$n_{e,\mathrm{NCD}}$ [n_{cr}]	SD material	N_p or N_d
1	-	64	-	CH_2	2.94×10^{11}
2	-	115	-	CH_2	3.84×10^{11}
4	20.46	115	1.06	CH_2	2.16×10^{11}
6	25.19	115	0.74	CH_2	1.96×10^{11}
7	18.19	115	0.74	CH_2	1.95×10^{11}
6b	25.19	115	0.74	CD_2	1.22×10^{11}

Brantov *L*

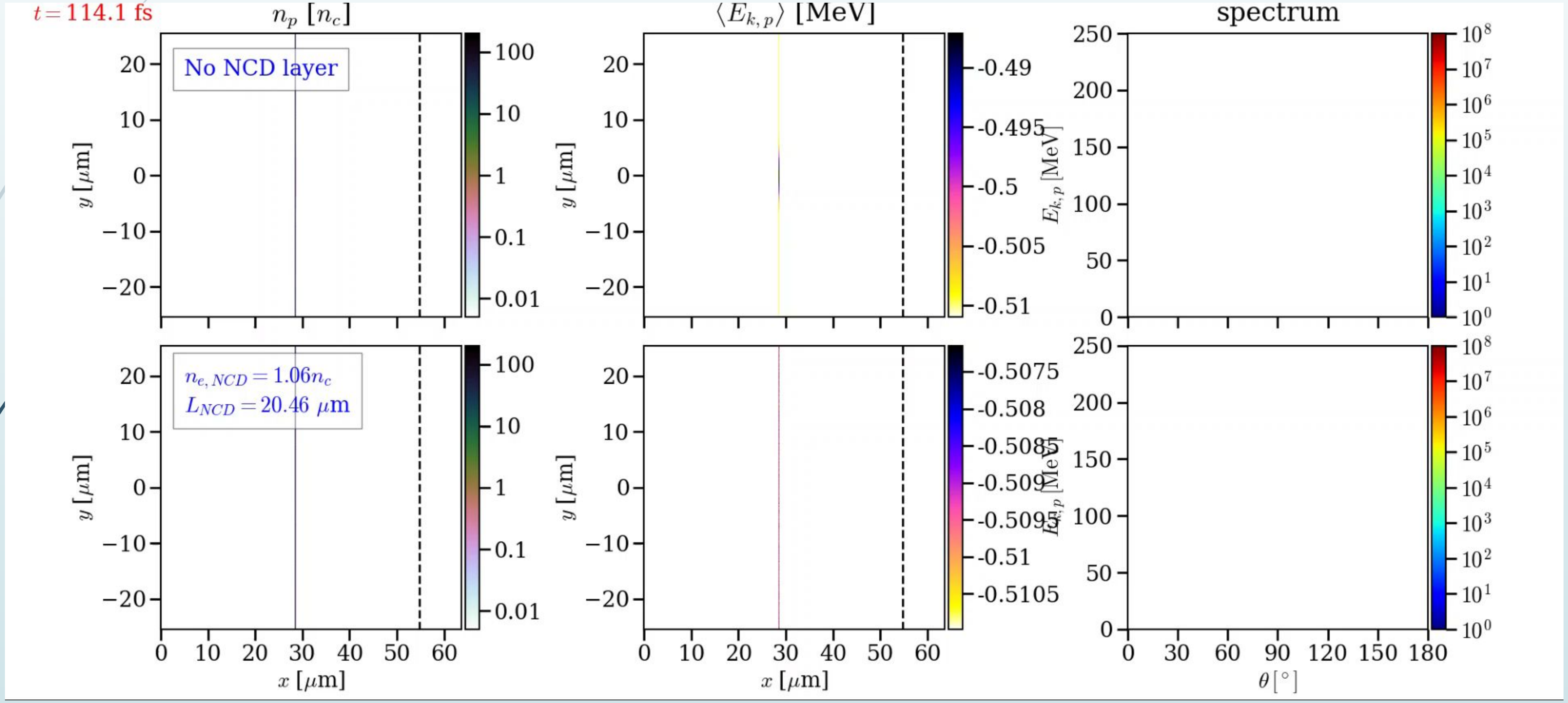
Other parameters are the same:

- Plasma: $n_{e,SD}$ = 200 n_{cr}
- Laser: D_0 = 5 µm, a_0 = 30.6, τ = 20 fs

Last column shows total number of accelerated protons or deuterons with energy above 1 MeV.



Proton acceleration with a single- and double-layer targets

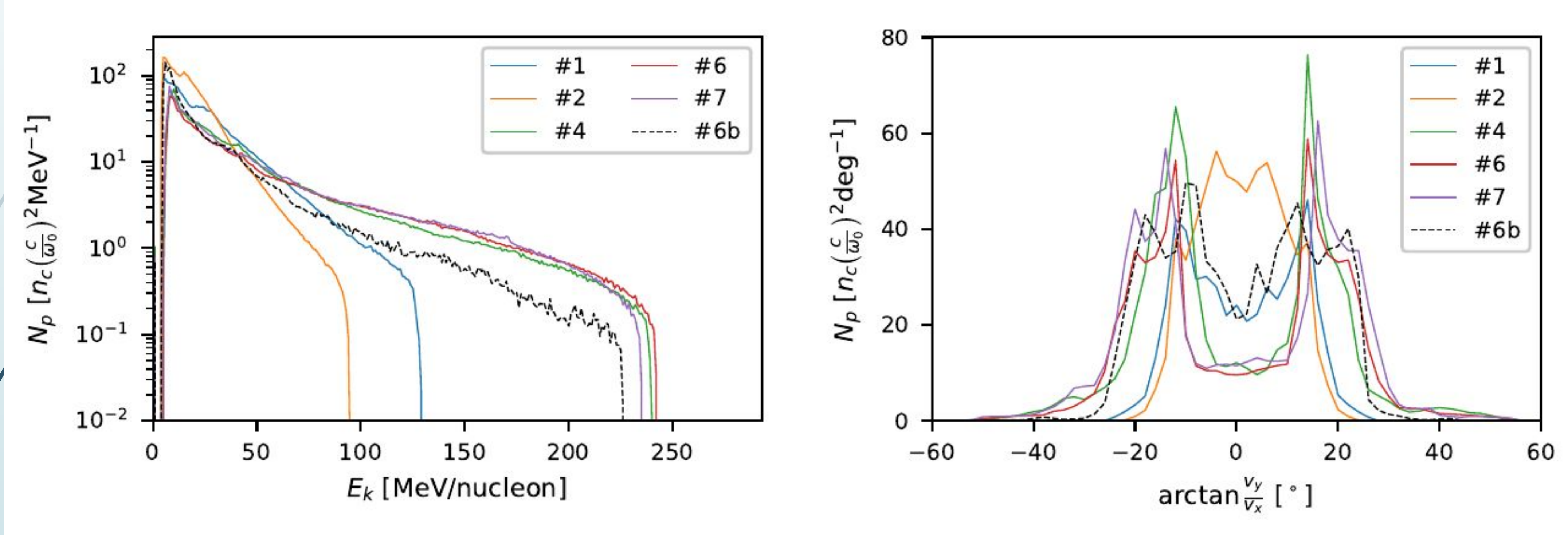


Case #2

Case #6

Tight laser self-focusing in near-critical plasma leads to significantly faster protons but with reduced number and larger angular divergence!

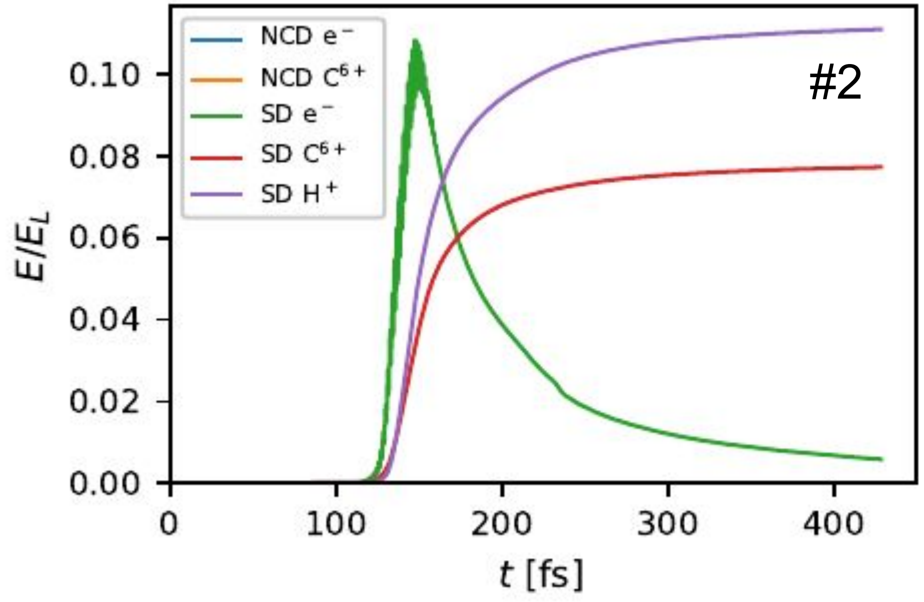
Ion acceleration with a single- and double-layer targets

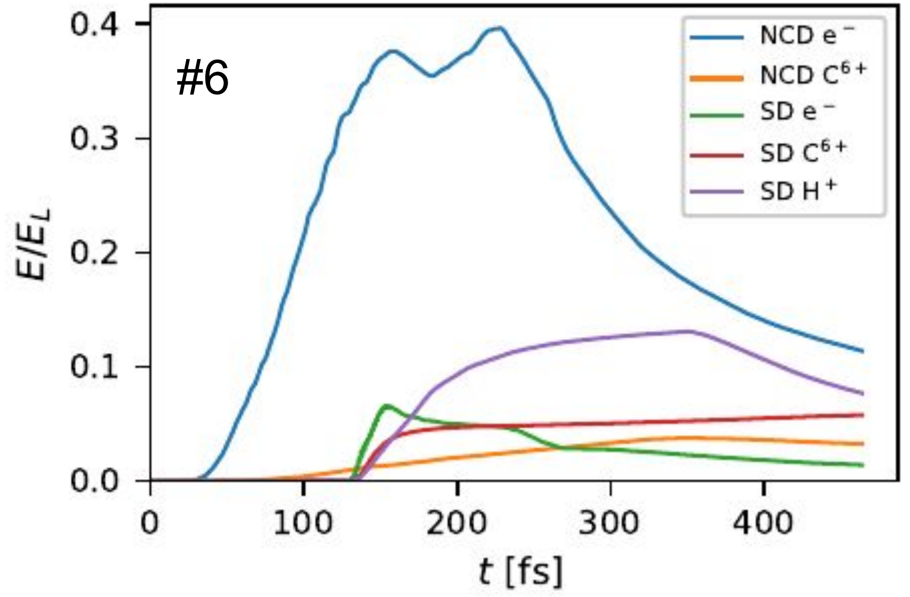


- NCD layer greatly enhances the final ion energy.
- NCD decreases the total ion number and worsens the divergence.

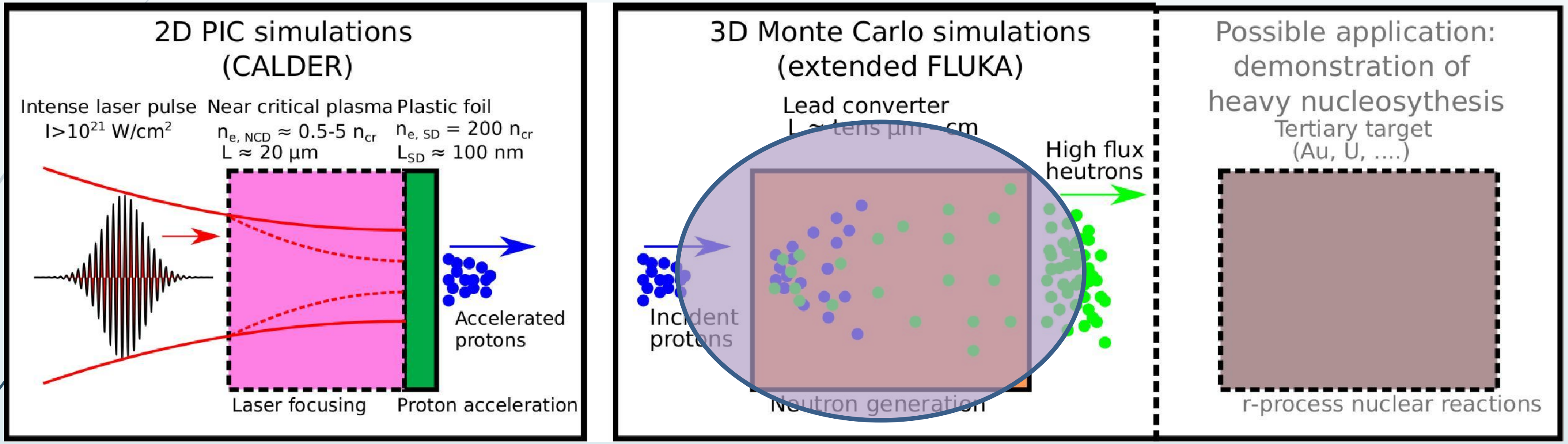
Laser energy conversion to particles

- Optimum near-critical plasma (NCP) can absorb over 40% of laser energy
- Fast electrons from NCP enhance proton acceleration from solid foil
- 10 − 12 % laser energy conversion into protons





Ultraintense-laser-based neutron generation



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- Apollon or ELI NP systems operate once per minute - it is rather a nuclear security concern.
- In fact, similar systems could be capable of running at 10 Hz.

Monte Carlo simulation with FLUKA

Extensions of the FLUKA code

- Loading of the primary particles from CALDER produced input
  - Transformation from 2D to 3D assuming the cylindrical symmetry
  - Each position (x,y) and momentum (p_x,p_y) rotated by a random azimuthal angle
  - Particle weight divided by the radial distance from the axis
- Implementation of the routine for the tracking of the particle flux temporal profile
  - Temporal profile considered both at the primary particles and all secondaries.

Simulation parameters

- Always at least 3.6×10^8 primary particles used.
- Convergence tests conducted up to 3.6×10^{10} primaries.
- Converter is a cylinder of r = 3 cm.

Further studies on the effect of the laser accelerated electrons and generated x-rays

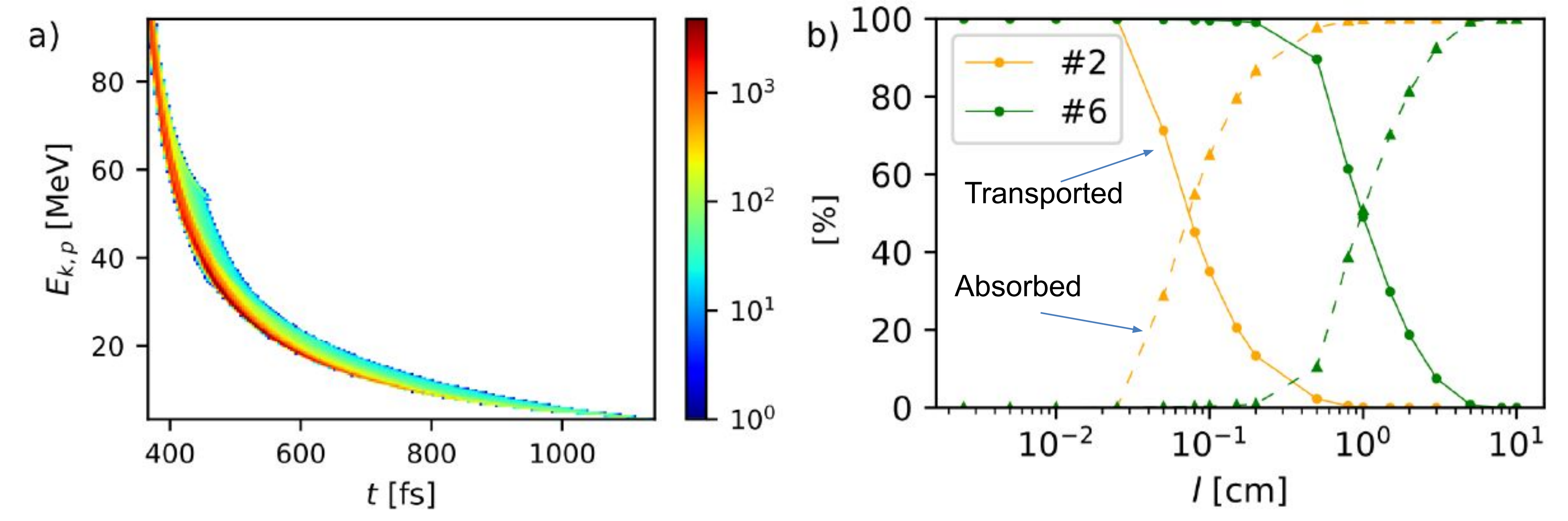
- their contribution to the final neutron yield is negligible.

Kudos to Bertrand Martinez for his contribution to the implementation of those routines.

Proton transport through Pb converter

Incident proton spectrogram #2

Monte Carlo simulation

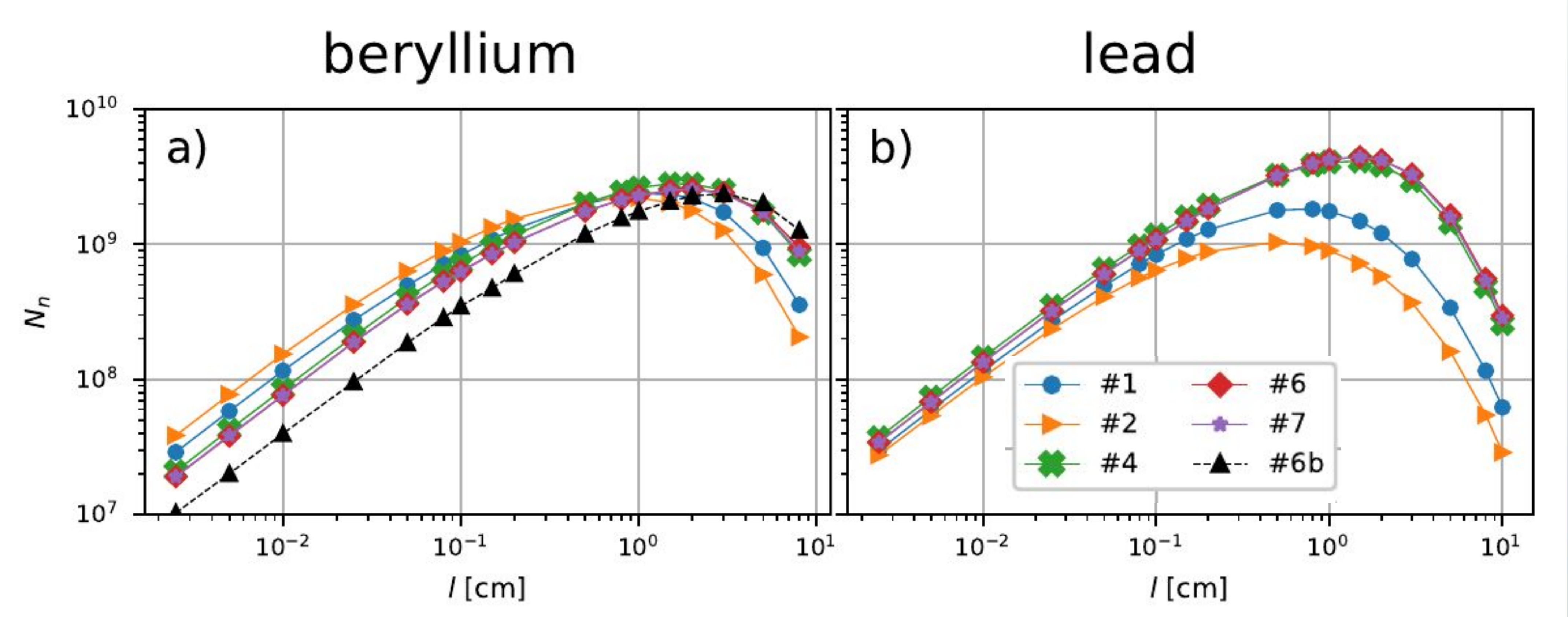


- Thin converters: $l \leq 250$ µm (#2) or 2 mm (#6), ≤ 1% of incident energy deposited
- Thick converters: $l > 0.5$ mm (#2) or 5 cm (#6), all incident energy deposited

Neutron source features

Total neutron number emitted from the rear side of the converter (i.e. in 2π sr)

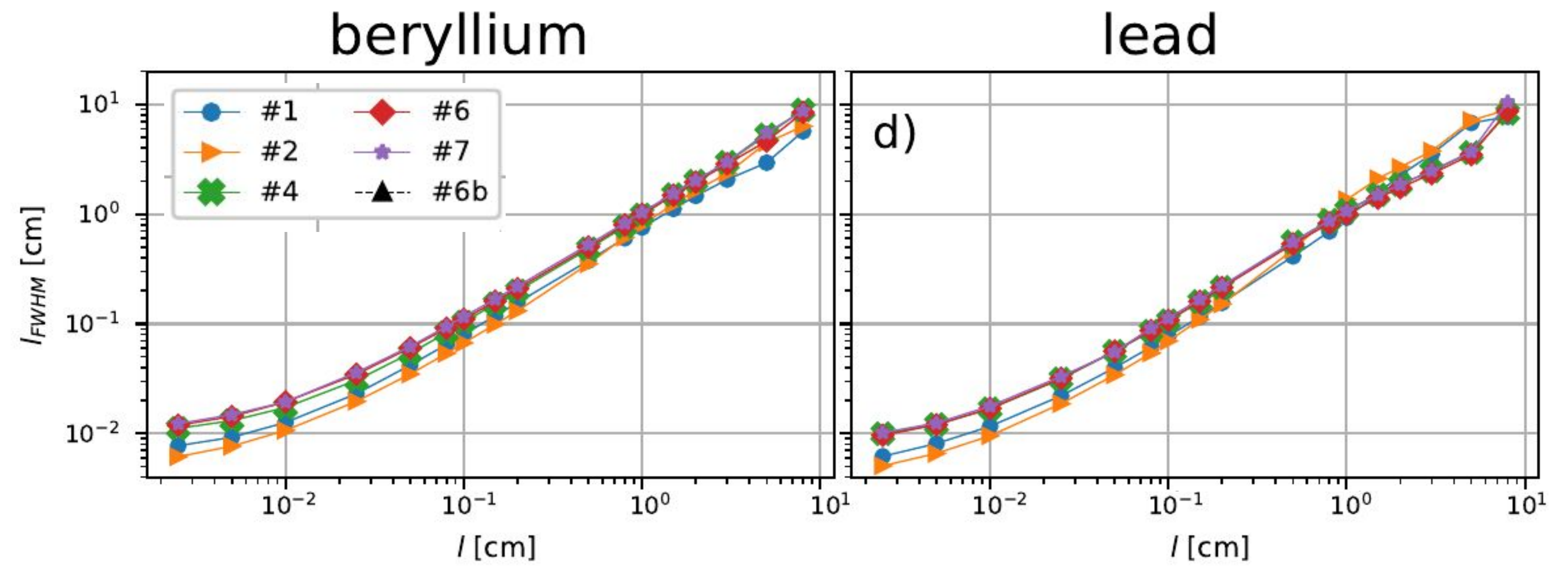
- Highest neutron production already in primary double layer target and lead converter.
- Only a weak dependence on the exact parameters of the front NCD layer.



Neutron source features

Neutron source FWHM at the rear side of the converter

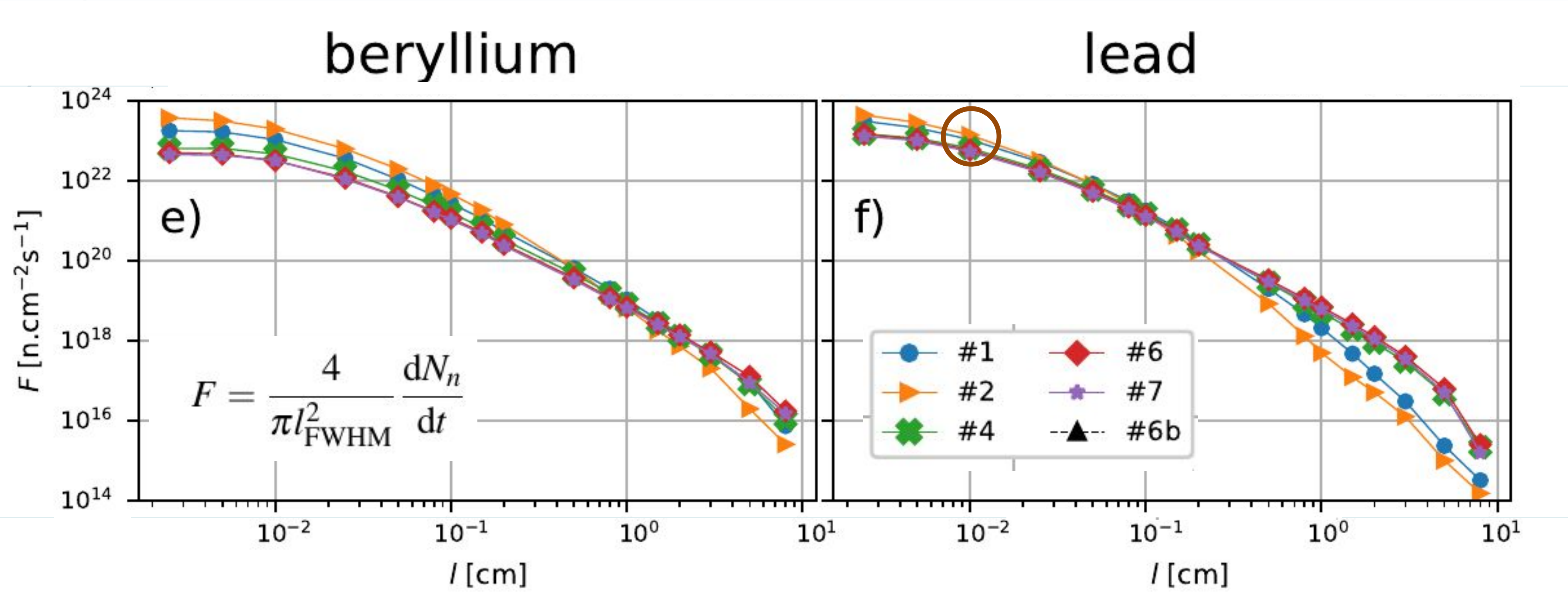
- The dependence $N_n(l, r)$ fitted by the Gaussian distribution.
- For converter thicknesses higher than ~0.5 mm source size depends linearly on *l*.



Neutron source features

The highest immediate neutron flux

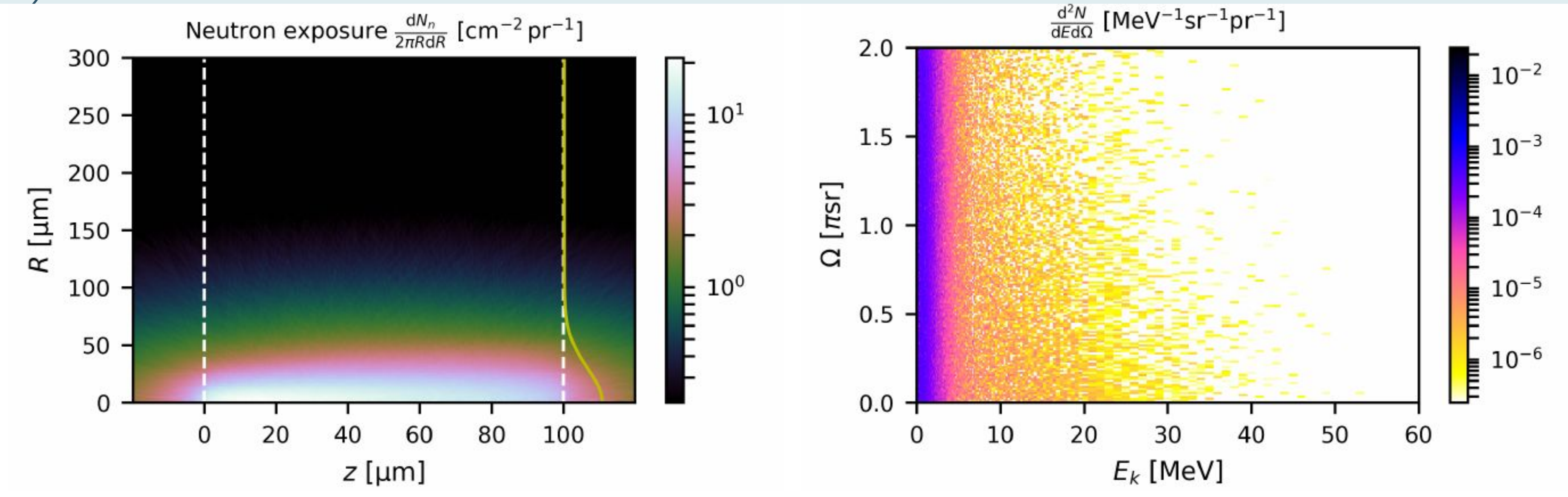
- Single layer proton acceleration targets in combination with thin converters can provide immediate flux above 10^{23} neutrons $cm^{-2}s^{-1}$.
- The strong dependence on the source size surpasses the relatively low neutron number.



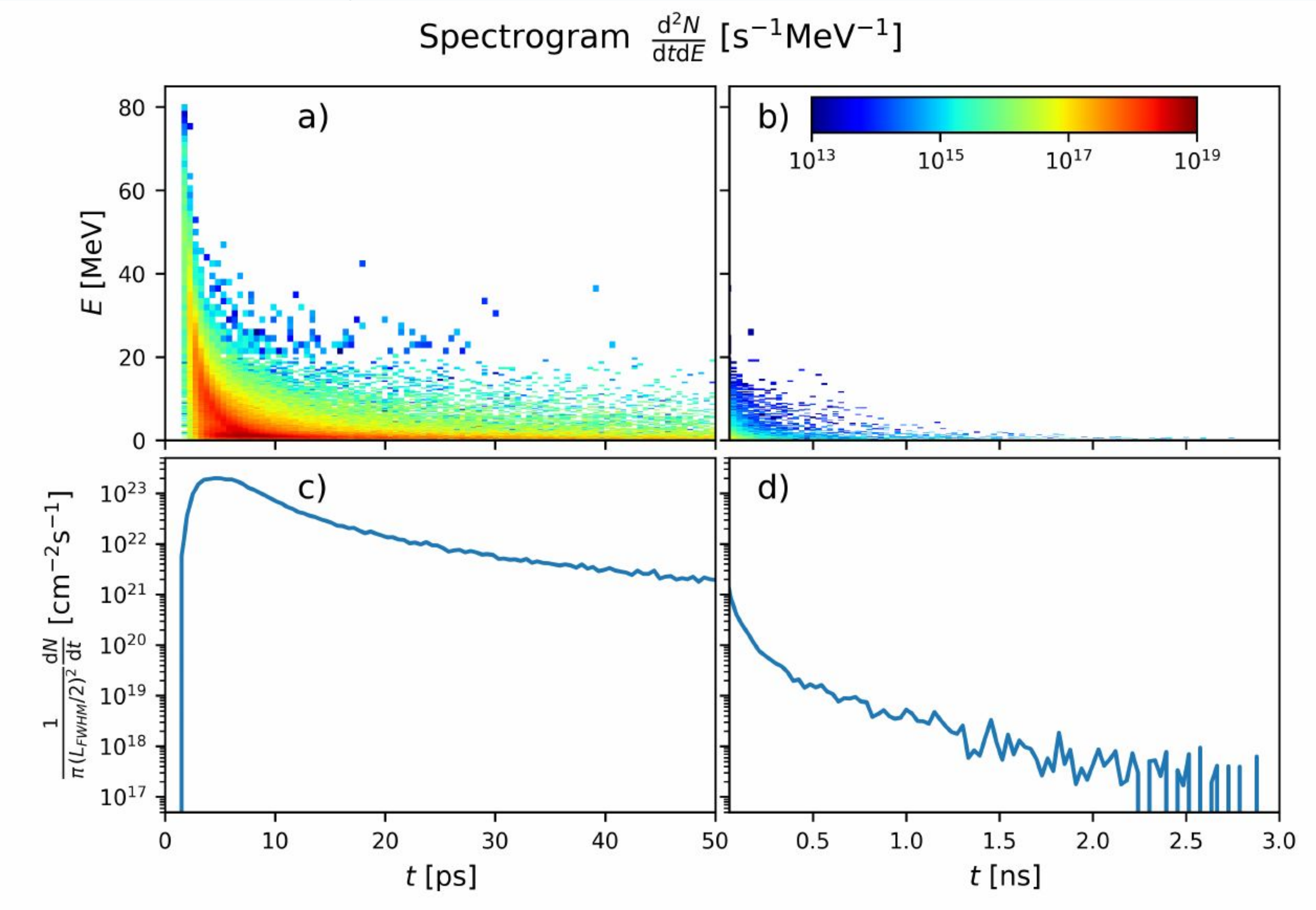
Neutron fluence and energy-angle spectra

Proton acceleration #2 + 100 μm thick Pb converter

- 1.54 × 10^{8} neutrons emitted per shot
- Transverse neutron source size ~ 95 μm
- Maximum immediate neutron flux ~ 1.41 × 10^{23} n $\mathrm{cm}^{-2}\,\mathrm{s}^{-1}$
- Maximum integrated neutron flux ~ 1.27 × 10^{6} n sr^{-1}



Neutron source features

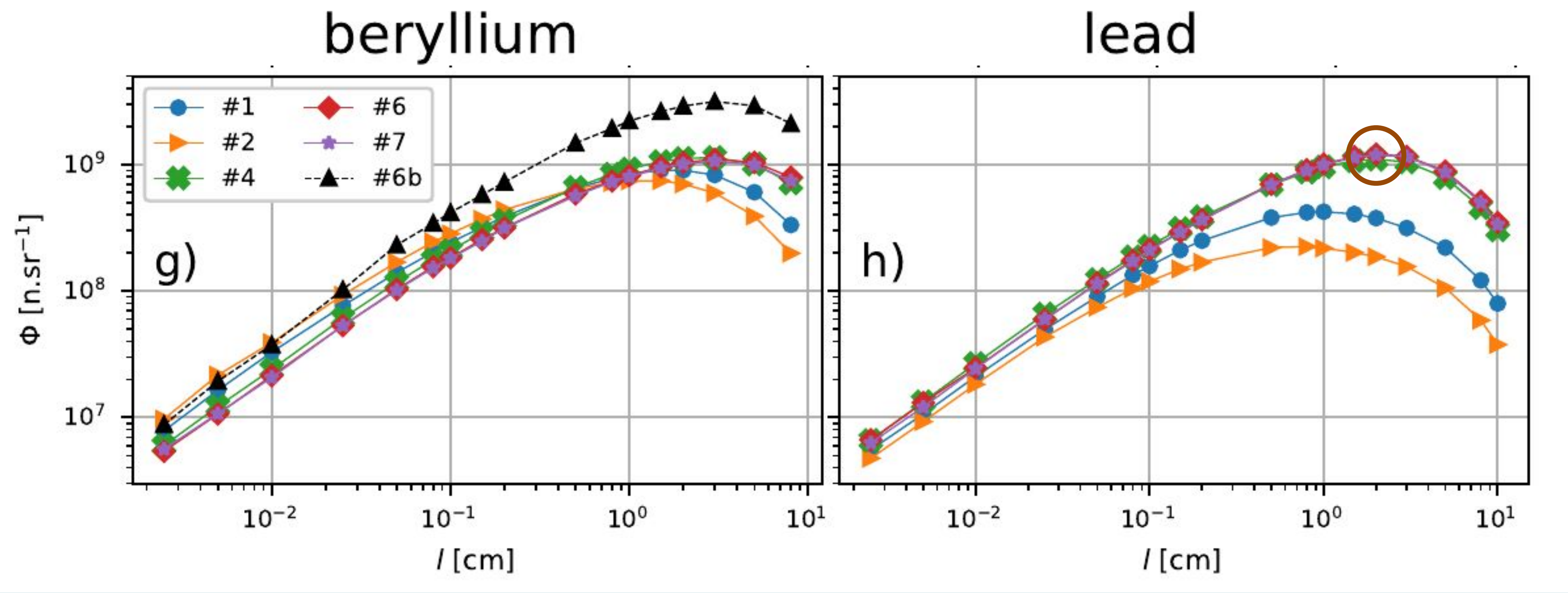


- A relatively high $N_n = 1.54 \times 10^8$
- $F = 1.98 \times 10^{23}$ neutrons cm^{-2}s^{-1}
- The most energetic neutrons are delivered within a few ps.
- After 1 ns, $F \approx 10^{19}$ neutrons cm^{-2}s^{-1},
- The less energetic neutrons continuously arrive.
- A flux higher than 10^{20} neutrons cm^{-2}s^{-1} is maintained for 240 ps in this case.

Neutron source features

The neutron flux at the axis: the quantity important for applications

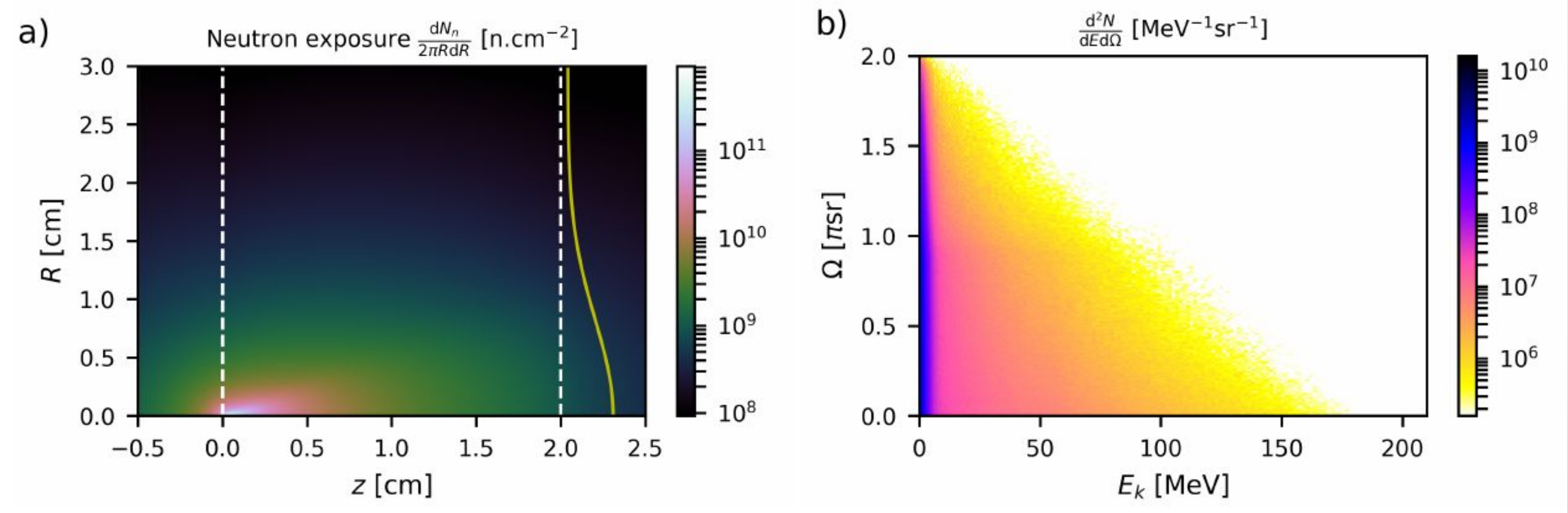
- In 1 PW regime, neutron generation is still slightly better for accelerated deuterons and a Be converter.
- The strong forward directionality of the nucleons released by the deuteron break-up.



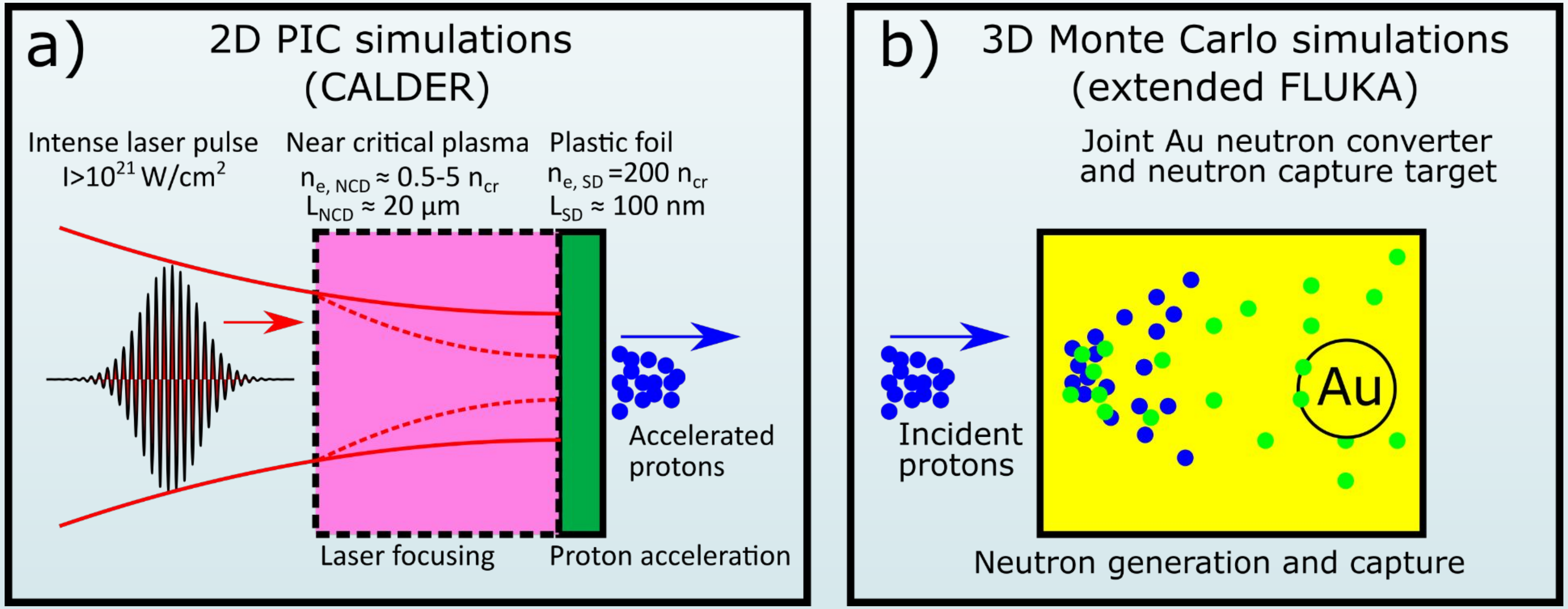
Neutron fluence and energy-angle spectra

Proton acceleration #6 + 2 cm thick Pb converter

- 4.25×10^9 neutrons emitted per shot
- Transverse neutron source size ~ 1.74 cm
- Maximum immediate neutron flux ~ 1.27×10^{18} n cm^{-2} s^{-1}
- Maximum integrated neutron flux ~ 8.40×10^7 n sr^{-1}

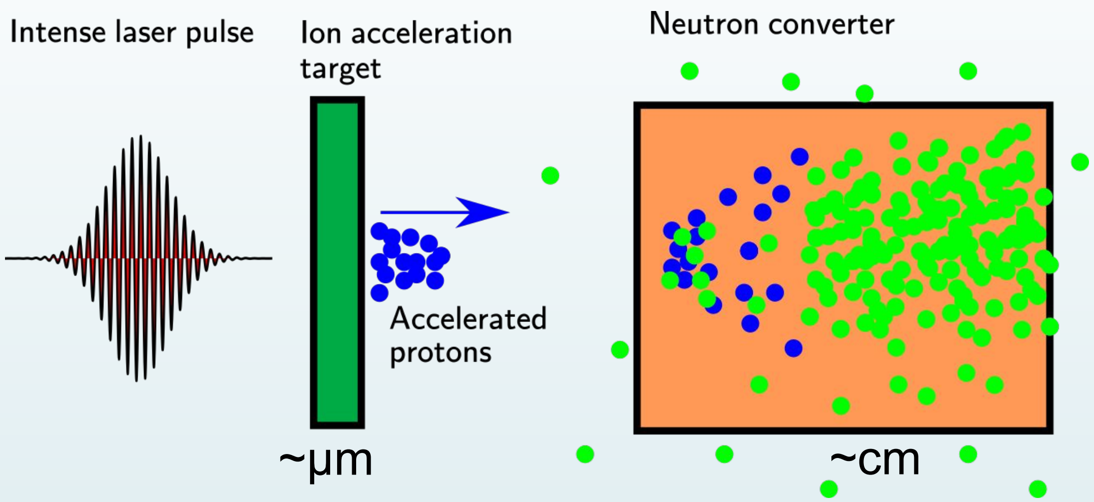


Simplified scheme for neutron capture



High Z material can be used for both neutron generation and capture with >20 MeV protons from advanced acceleration schemes.

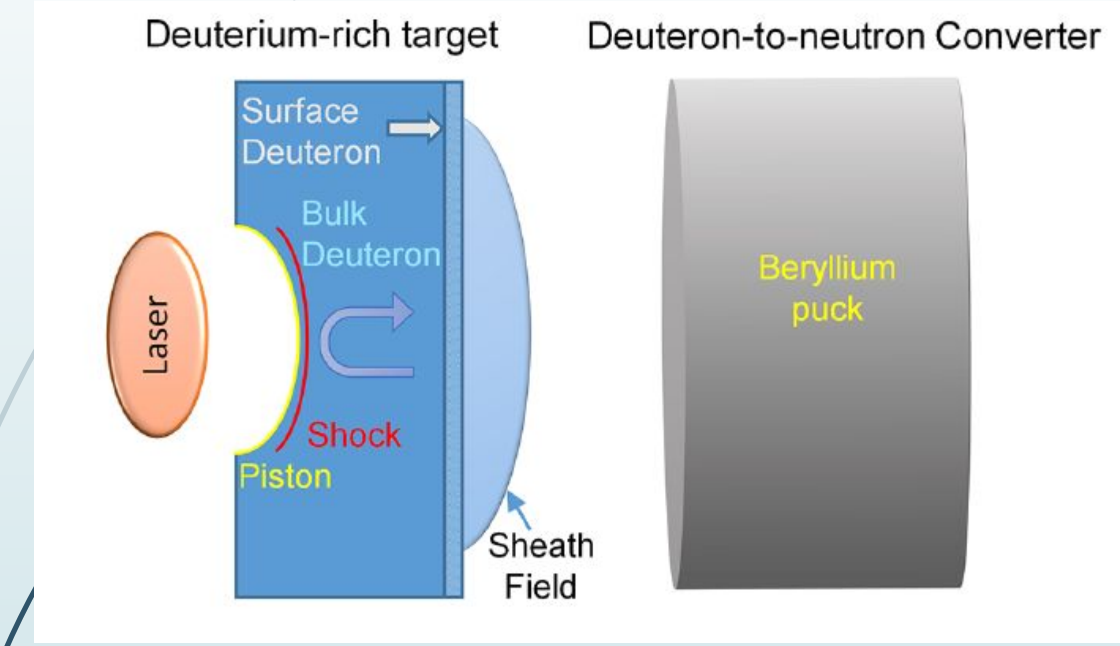
Laser-driven neutron source

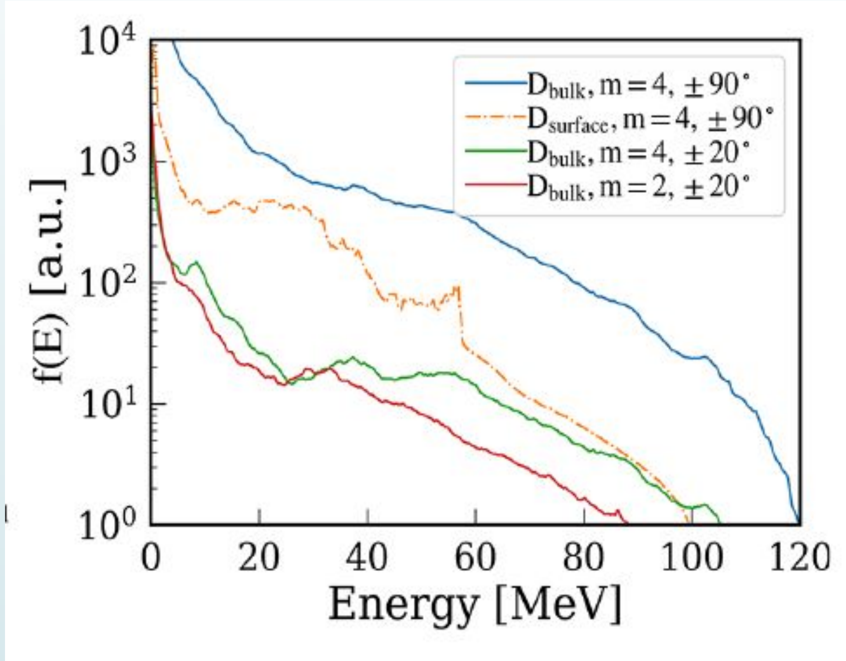


State-of-the-art results achieved at PHELIX@GSI, TRIDENT@LANL or LFEX@ILE Osaka.

	E [J]	τ[fs]	f		F [N/sr]	F/E[N/sr/J]	fF [N/sr/s]
Roth, et al. PRL (2013)	80	600	once per hour	CD_2+Be	4.40×10^9	5.50×10^7	1.22×10^6
Kleinschmidt et al., Phys. Plasmas (2018)	150	500	once per 90 minutes	CD_2+Be	1.42×10^{10}	9.47×10^7	2.63×10^6
Huang et al., APL (2022) –theoretical suggestion	282	1000	once per hour	CD2+Be	1.70×10^{11}	6.03×10^8	4.72×10^7
Günther et al., Nat Com. (2022)	20	700	once per hour	CHO+Au	4.93×10^9	2.47×10^8	1.37×10^6
Mirfayzi et al., SciRep (2020)	300	1200	once per hour	CD_2+Be+PET+H	8×10^5 cold		
Horný et al., SciRep (2022)	22	20	once per minute	CH_2+Pb	1.2×10^9	5.5×10^7	2.0×10^7

LANL shock acceleration design





- Collisionless shock shifts the deuteron spectrum towards higher energies.
- In principle, no need of inclusion of vacuum between target and converter.
- Efficient collisionless shock acceleration yet to be demonstrated.

Huang, C. K., Broughton, D. P., Palaniyappan, S., Junghans, A., Iliev, M., Batha, S. H., ... & Favalli, A. (2022). High-yield and high-angular-fluence neutron generation from deuterons accelerated by laser-driven collisionless shock. *Applied Physics Letters*, *120*(2), 024102.

Neutron-rich isotopes abundance model

Based on Zagrebaev, V. I., et al. PRC 84.4 (2011): 044617.

Abundance of (A+i)-th isotope

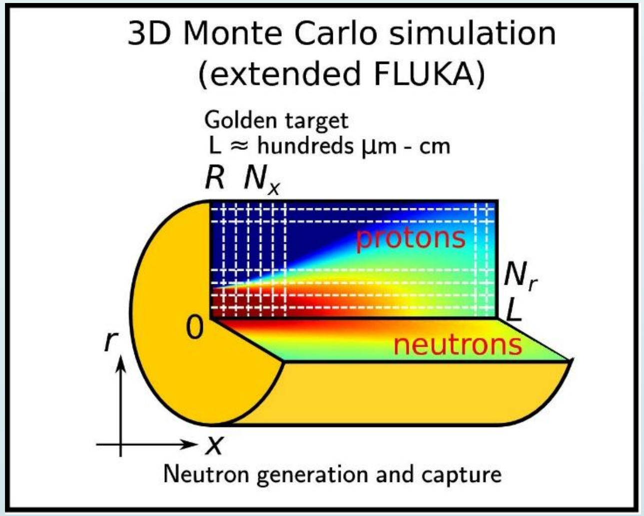
$$b_1 = \frac{N_1}{N_0^0} = \mu_1 \exp(-\mu_1) \simeq \mu_1,$$

$$b_2 = \frac{N_2}{N_0^0} = \frac{\mu_1 \mu_2}{2} \exp(-\mu_2) \simeq \frac{\mu_1 \mu_2}{2}$$

neutron areal density

neutron capture cross-section

$$\boxed{\mu_i = \frac{\mathrm{d}N_n}{\mathrm{d}S} \sigma_i}$$


3D Monte Carlo simulation (extended FLUKA)

Generalisation: radially and energy dependent flux

$$\mu_i(x, r) = \int \frac{\mathrm{d}^2 N_n}{\mathrm{d}S \mathrm{d}E}(x, r, E) \sigma_i(E) \mathrm{d}E.$$

can be taken e.g. from FLUKA simulation

- Number of (A+2) isotope nuclei scales with
  - Square of the neutron flux → **spatially small sources**
  - Square of the neutron capture cross-section → **slower than epithermal neutrons**

Neutron-rich isotopes calculation

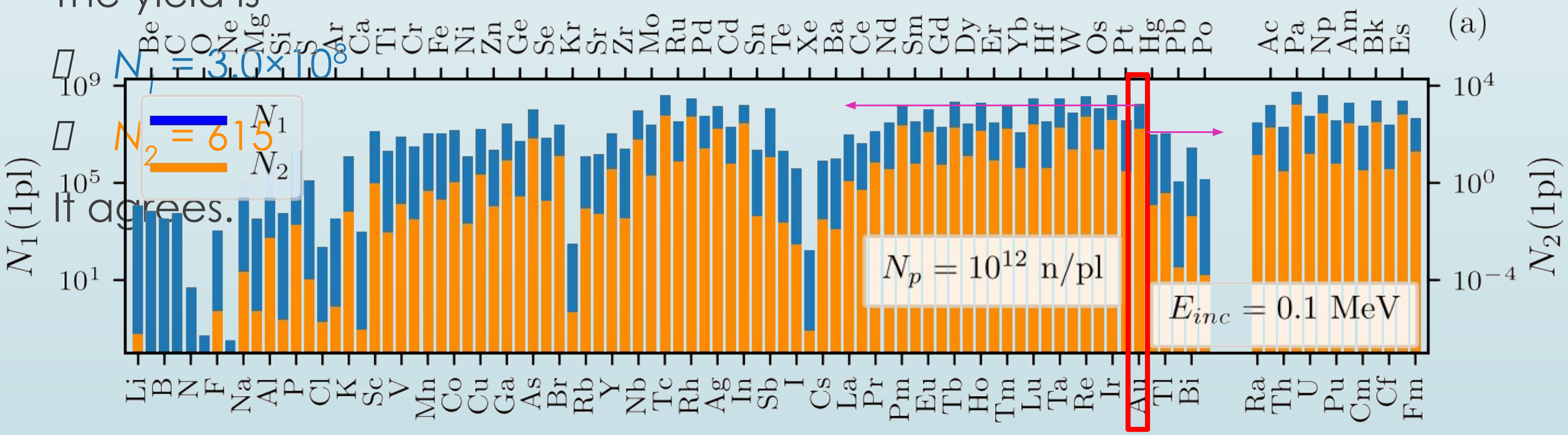
Calibration: Hill & Wu parameters

- 100 keV neutrons
- N_n = 1.0e12
- No divergence!
- S = 25 um^2
- Au converter, l = 100 um

The yield is

- $N_1 = 3.0\times10^8$
- $N_2 = 615$

It agrees.

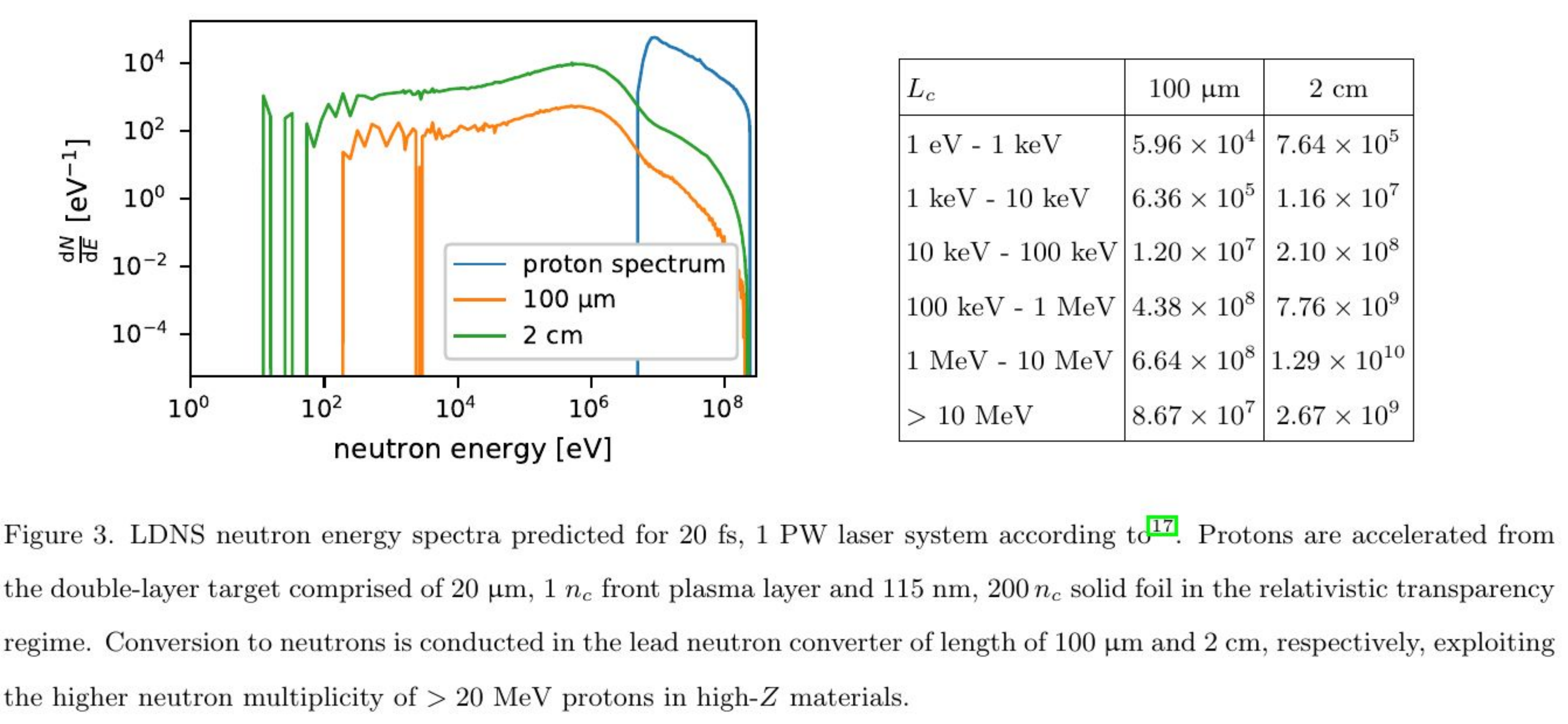


Implementation details:

- Written Python
- Neutron cross sections from TENDL-2019 library.

Laser-driven neutron source

Optimised proton and neutron spectra predicted for Apollon@1 PW



L_c	100 μm	2 cm
1 eV - 1 keV	5.96×10^4	7.64×10^5
1 keV - 10 keV	6.36×10^5	1.16×10^7
10 keV - 100 keV	1.20×10^7	2.10×10^8
100 keV - 1 MeV	4.38×10^8	7.76×10^9
1 MeV - 10 MeV	6.64×10^8	1.29×10^{10}
> 10 MeV	8.67×10^7	2.67×10^9

Figure 3. LDNS neutron energy spectra predicted for 20 fs, 1 PW laser system according to[17]. Protons are accelerated from the double-layer target comprised of 20 μm, 1 n_c front plasma layer and 115 nm, 200 n_c solid foil in the relativistic transparency regime. Conversion to neutrons is conducted in the lead neutron converter of length of 100 μm and 2 cm, respectively, exploiting the higher neutron multiplicity of > 20 MeV protons in high-Z materials.

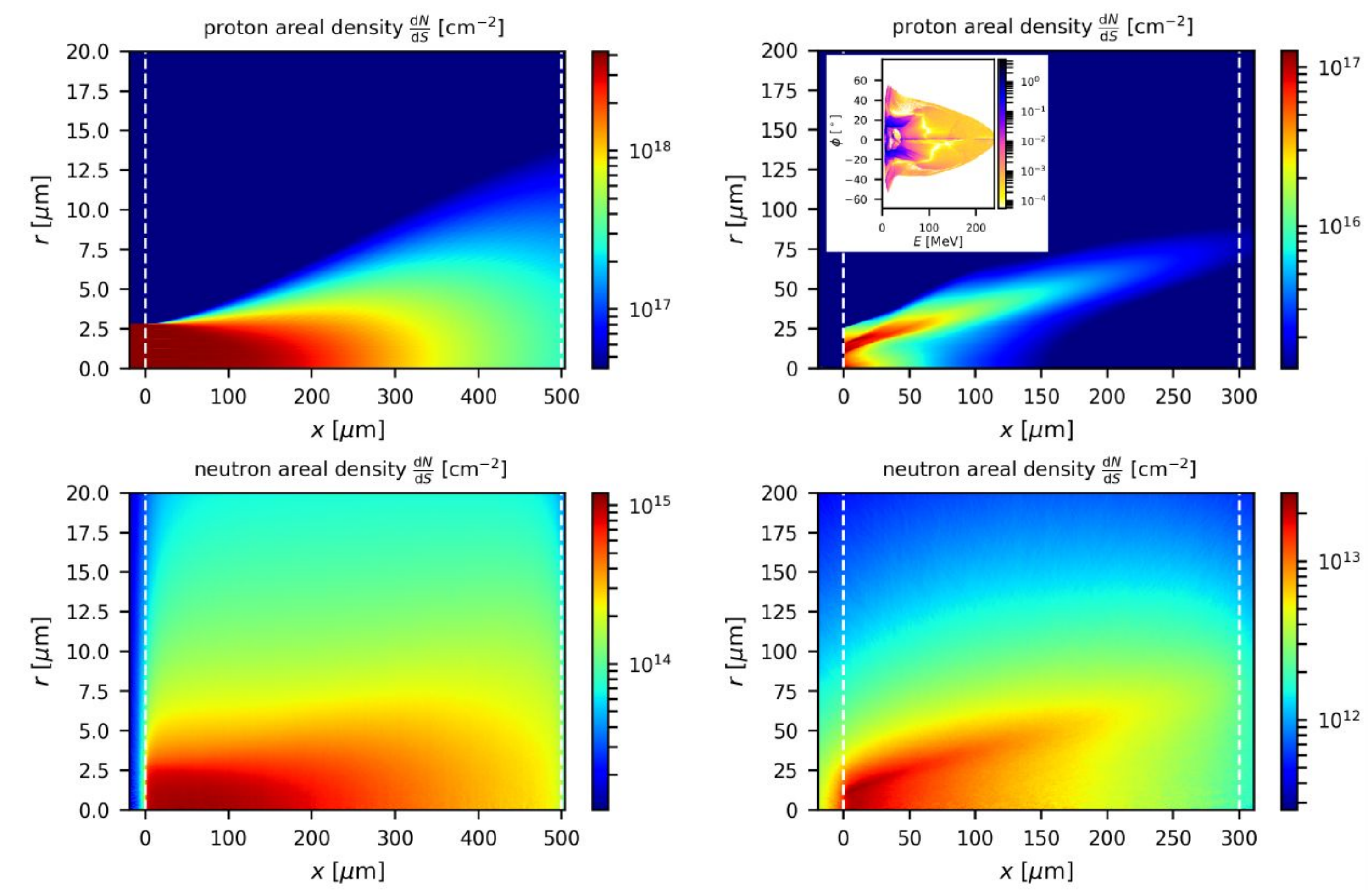
Neutron source size is comparable to converter length!

Horný, V. et al. (2022). High-flux neutron generation by laser-accelerated ions from single- and double-layer targets. *Scientific Reports, 12,*

Proton transport and neutron generation

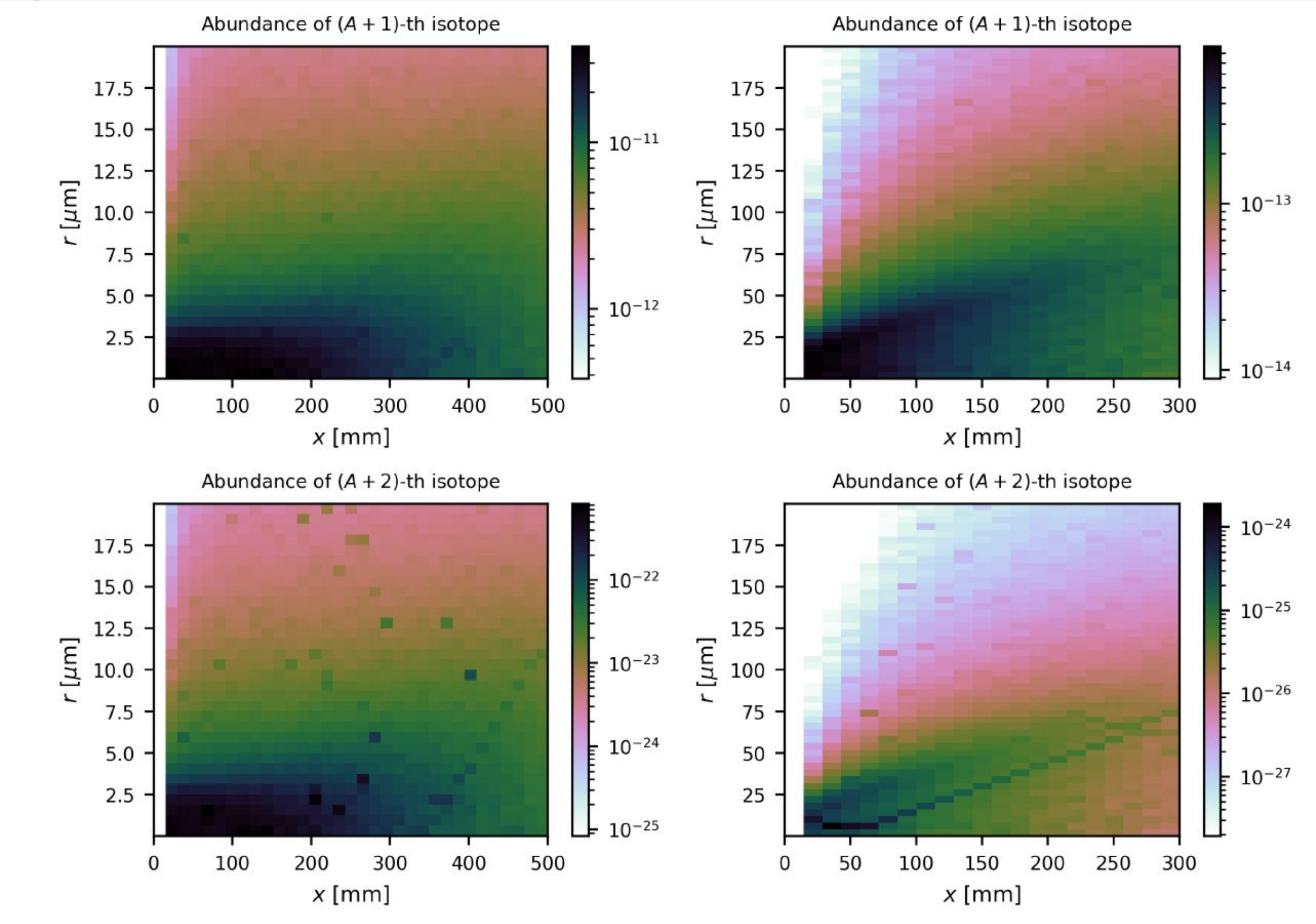
10^{12} protons of E = 100 MeV

simulated double layer case



Neutron-rich isotopes generation

10^{12} protons of E = 100 MeV simulated double layer case



Generated isotopes

10^{12} protons of E = 100 MeV

- $N_1 = 1.8\times10^{5}$
- $N_2 = 7.3\times10^{-7}$

Double layer primary target

- $N_1 = 4.7\times10^{5}$
- $N_2 = 8.8\times10^{-8}$

Orders of magnitude lower than with idealized neutron source from literature.

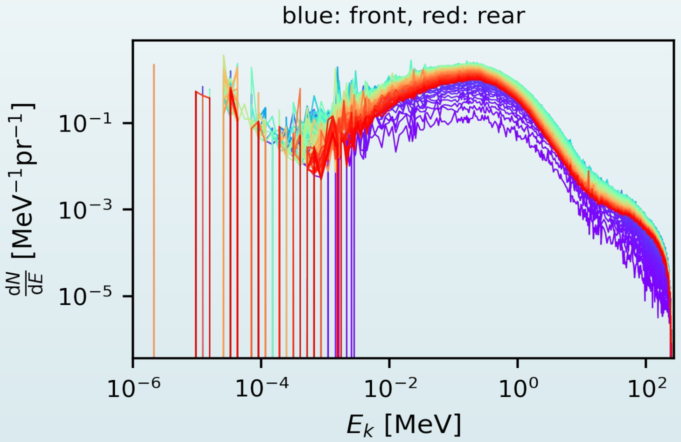
Neutron-rich isotopes calculation

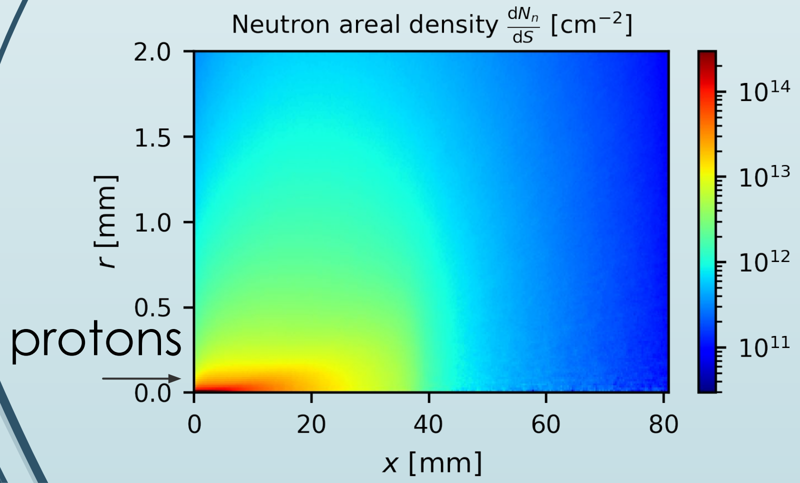
Scholar case: spallation of 1 GeV protons in a golden bulk

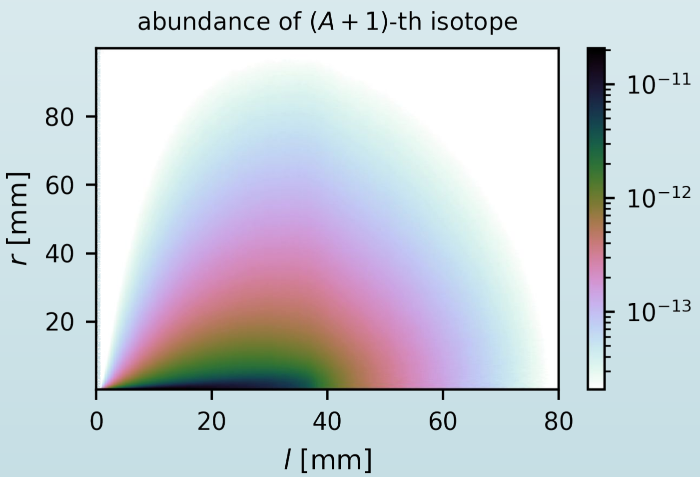
- Proton beam area, S = 25 um^2
- $N_p = 1.0\times10^{13}$
- Au converter, L = 8 cm

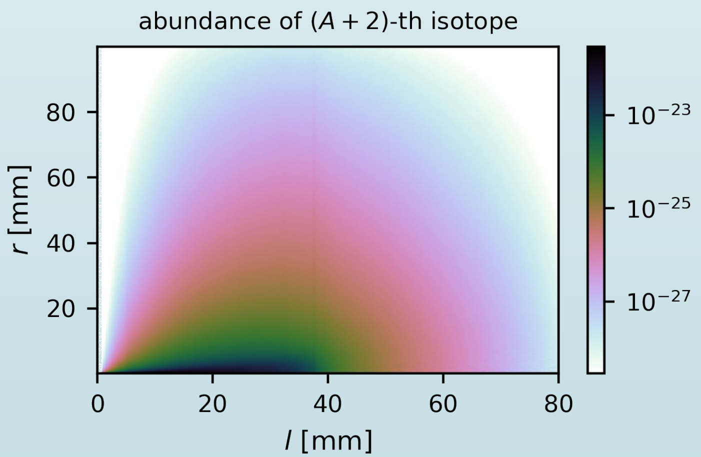
- $N_1 = 9.5\times10^{12}$
- $N_2 = 4.6$

Neutron spectrum


blue: front, red: rear


Neutron areal density $\frac{dN_n}{dS}$ [cm^{-2}]


abundance of $(A+1)$-th isotope


abundance of $(A+2)$-th isotope

Neutron-rich isotopes calculation

Mirfayzi, S. R., et al., Scientific Reports 2020 10(1), 1–8.

Standard LDNS

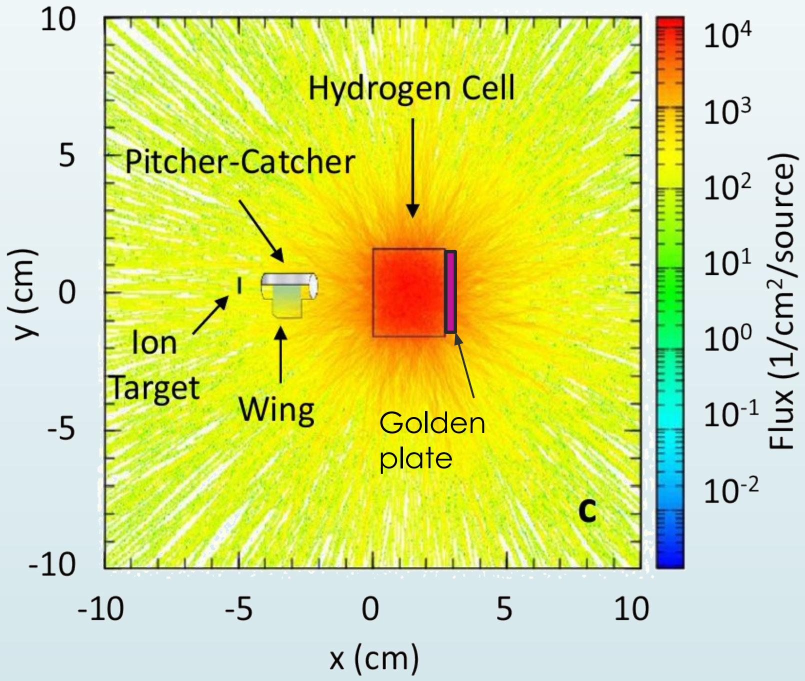
- TNSA with 300 J, 1.2 ps system and 5 µm C_2D_4 target.
- Be converter
- Fast neutron flux 1×10^9 n/sr.

Two stage moderation

- PET pre-moderator attached to converter
- Cryogenic H_2 moderator

Let us assume

- 100 Hz, 8 hours operation
- Cold neutrons , E_C = 20 meV
- 1×10^7 cold neutrons per shot
- total integrated flux at the moderator surface: 1×10^{12} n/cm^2



Discussion

What was ignored?

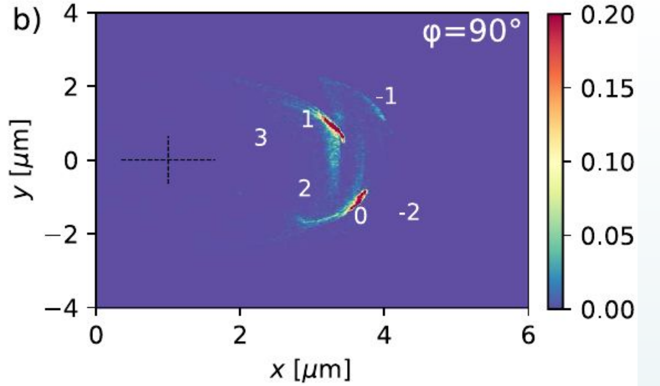
- Decay of (A+1)-isotopes.
  - ^{198}Au half-life is 2.7 days.
- Fact that ^{199}Au decay is hardly observable.
- Potential improving effect
  - (A+1)-isotopes are for up to 1 ns in excited state
  - neutron capture cross-section might be higher

Conclusion

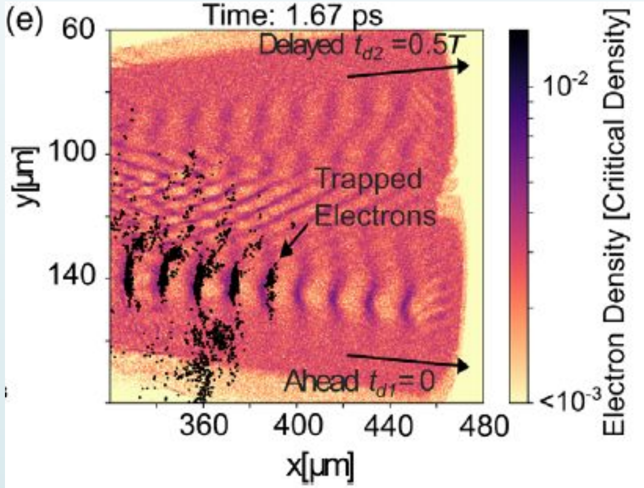
- Recent literature overestimate the potential of the laboratory studies of astrophysical *r*-process.
- Generation of neutron-rich (≥A+2) isotopes at laser systems in principle possible, but makes no sense.

Advertisement

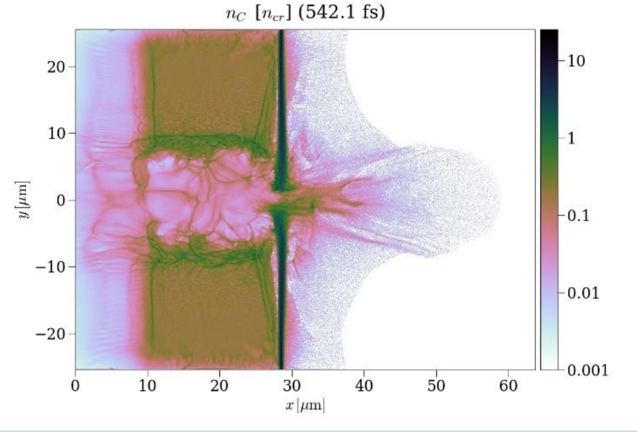
Horný, V., & Veisz, L. (2021). Generation of single attosecond relativistic electron bunch from intense laser interaction with a nanosphere. *Plasma Physics and Controlled Fusion*, *63*(12), 125025.



Chen, Q., Mašlárová, D., Wang, J., Li, S. X., Horný, V. & Umstadter D. (2022). Transient relativistic plasma grating to tailor high-power laser fields, wakefield plasma waves, and electron injection. *Physical Review Letters*, 128, 164801.



Horný, V., Chen, S.N., Davoine, X., Lelasseux, V., Gremillet, L. & Fuchs, J. (2022). High-flux neutron generation by laser-accelerated ions from single- and double-layer targets. Scientific Reports, 12, 19767



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