Investigating the Solid Deuterium in the PSI UCN Source Moderator

PSI LTP Seminar, 24.9.2018

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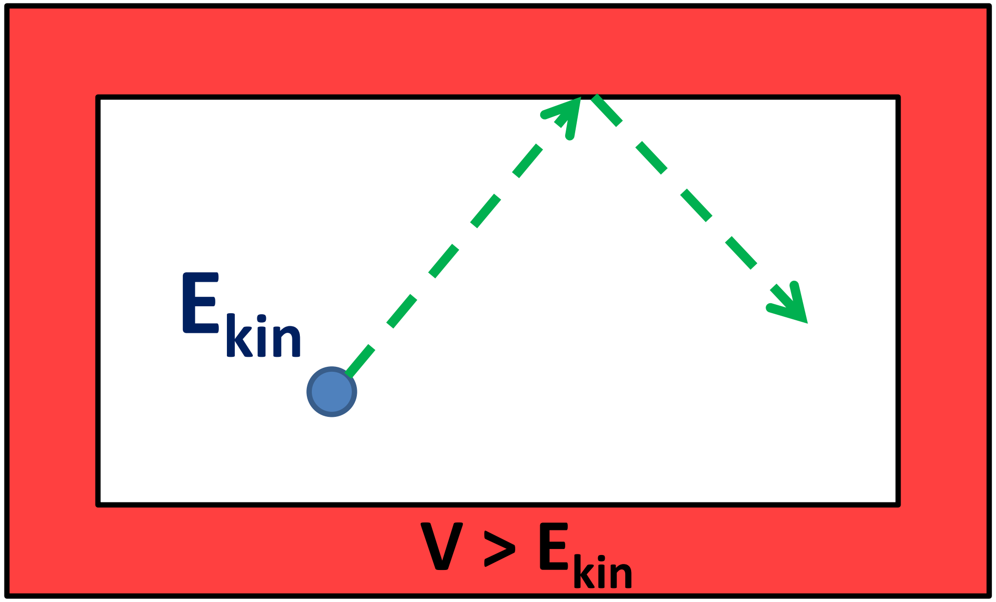
Paul Scherrer Institute, Switzerland

- What are ultracold neutrons (UCN) and their uses?
- Working principle of the UCN source at the Paul Scherrer Institute (PSI)
- Investigation and characterization of the behavior of the D_2 used in the PSI UCN source

What Are UCN?

- **Ultracold Neutrons (UCN): Are totally reflected on neutron optical potential, storage possible** in vessels made of appropriate materials for a timespan of several minutes limited by their β-decay lifetime (≈ 15 minutes)
- very slow neutrons, typically classified as having a **kinetic energy of ≤ 335 neV** (8 m s^{-1} , 3 mK)

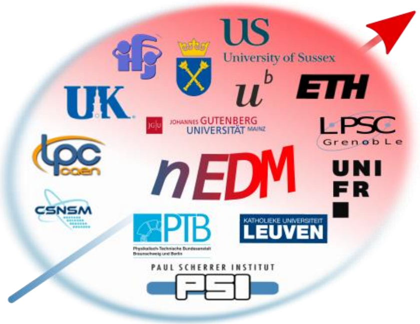


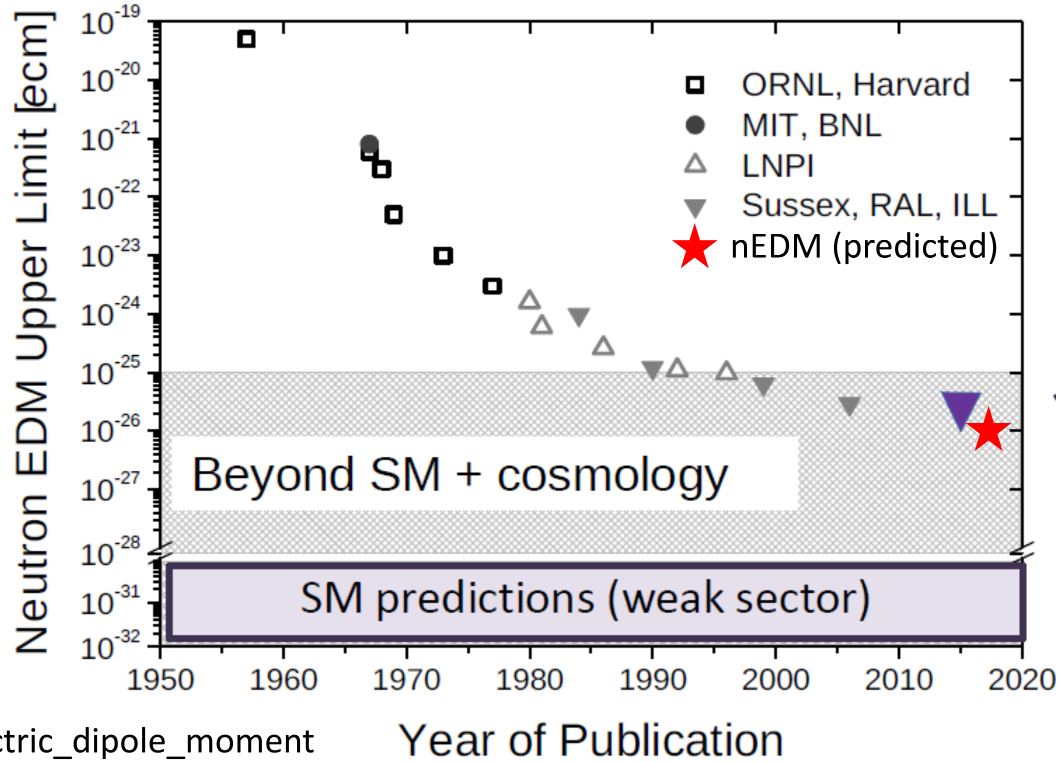
Material	V [neV]
^{58}Ni	335
Fe	210
Cu	168
Al	54
Ti	-48

Research with UCN

- **Valuable tools in high precision physics experiments,** e.g. measurements of free neutron lifetime and neutron electric dipole moment (nedm), for example the **nEDM (dismantled in Oct 2017) and future n2EDM experiments at PSI**
- **Precision** in experiments using UCN typically **scales with $\sqrt{N}$ → high output desired**

Evolution of the nedm limit

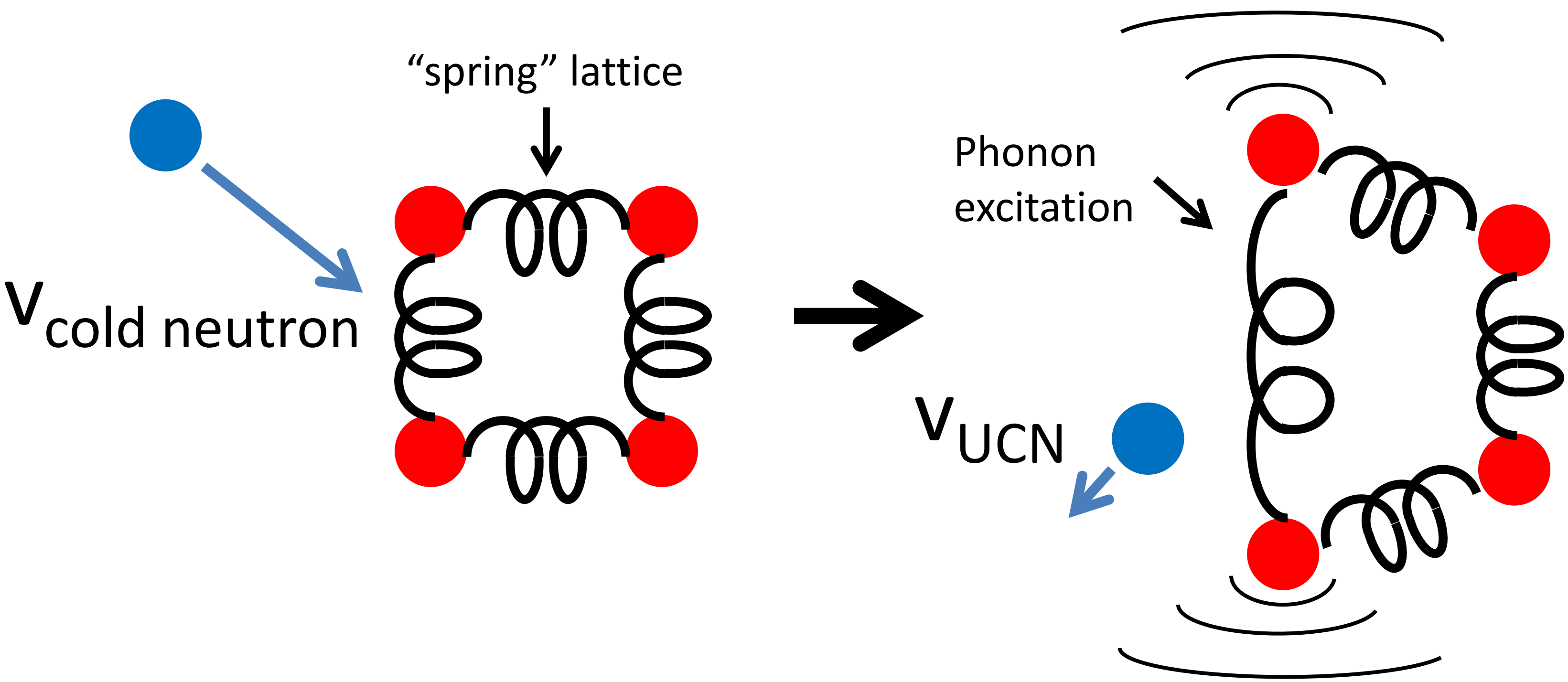




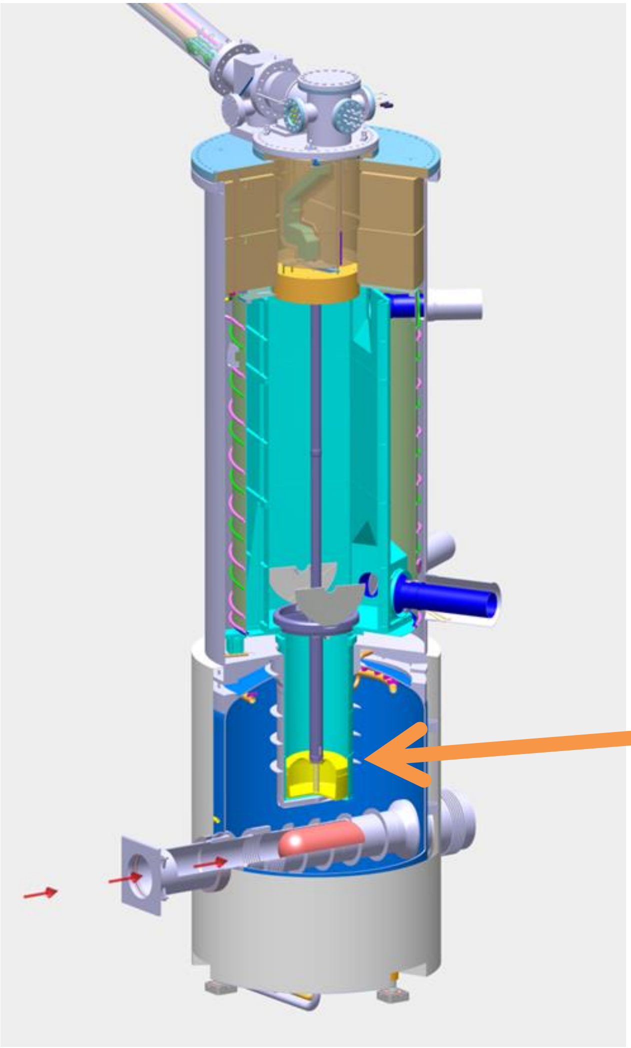
Plot originally by Andreas Knecht, modified,
https://en.wikipedia.org/wiki/Neutron_electric_dipole_moment

Production of UCN at PSI

- **Cold or thermal neutrons have the possibility to transfer nearly all of their kinetic energy through phonon excitation in solid deuterium (sD_2)**
- **Achieve higher UCN densities than the actual Maxwell-Boltzmann distribution** at the temperature of the D_2 would be → **“superthermal” production**

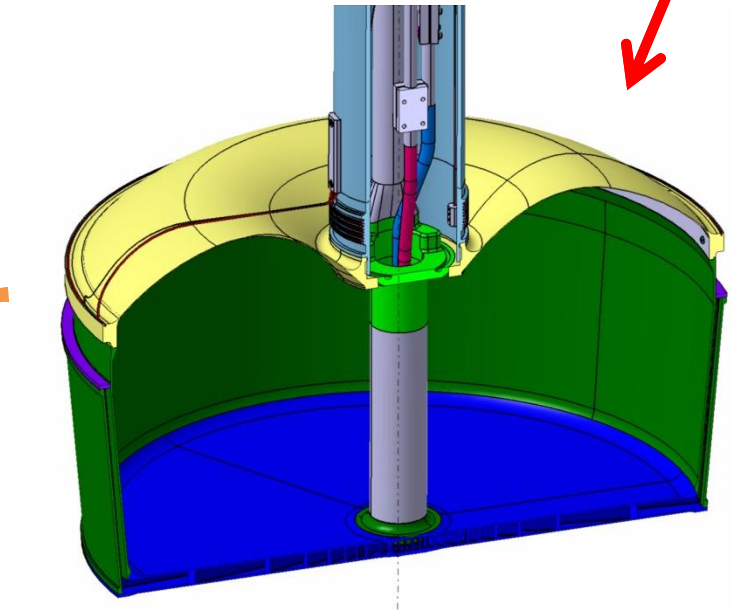


PSI UCN Source: Working Principle



Heart of the UCN source:

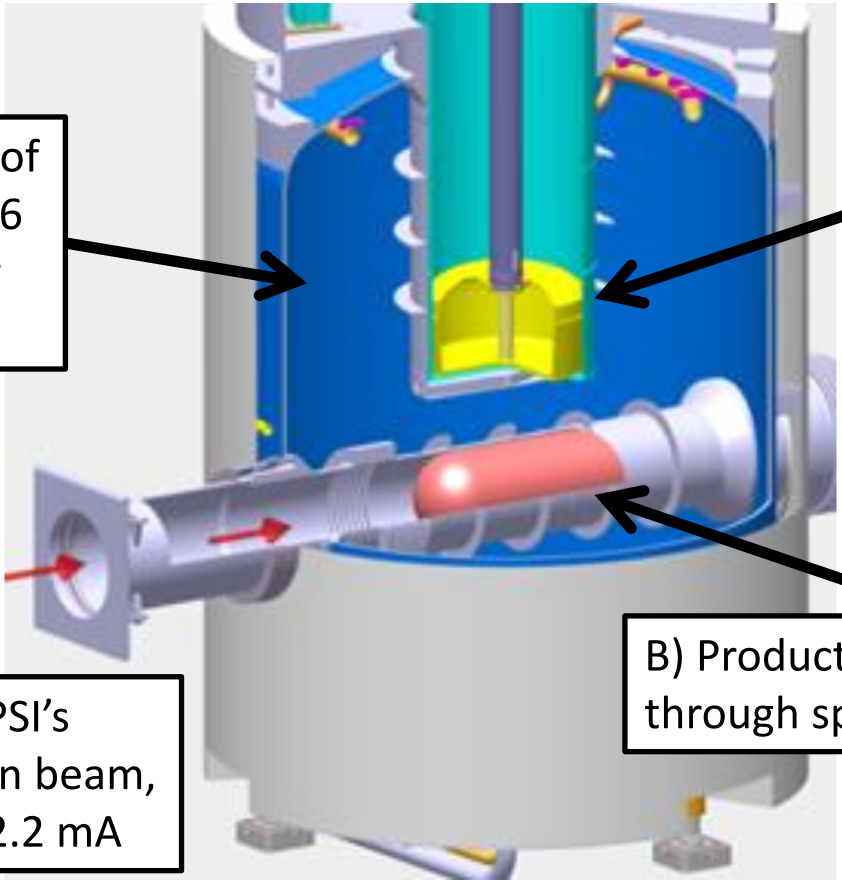
- Moderator vessel filled with solid D_2 at 5 K
- Closed system, no visual inspection possible, important later on



First Step: Pulsed UCN Production

C) Thermalization of fast neutrons in 3.6 m^3 of heavy water (D_2O) at 300 K

D) Fraction of incoming thermal neutrons is moderated towards cold regime and subsequently down-scattered into UCN regime using solid D_2 at 5 K



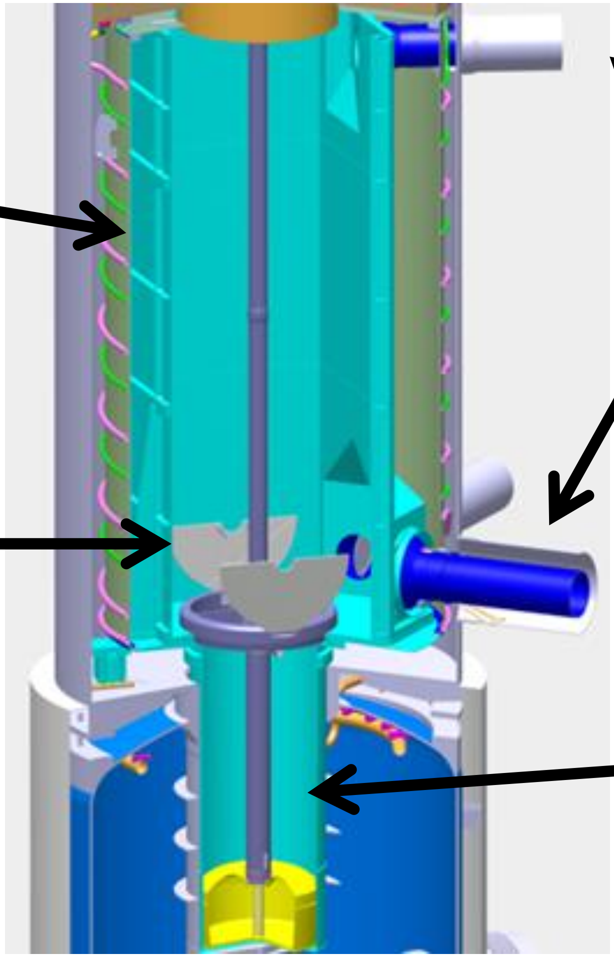
A) Pulse (8 s) from PSI's high intensity proton beam, 1.3 MW, 590 MeV, 2.2 mA

B) Production of fast neutrons through spallation in Pb target

Second Step: Storage and Extraction

Intermediate DLC-coated storage volume, 2 m^3

Flaps to close off storage tank at end of UCN production, i.e. proton pulse

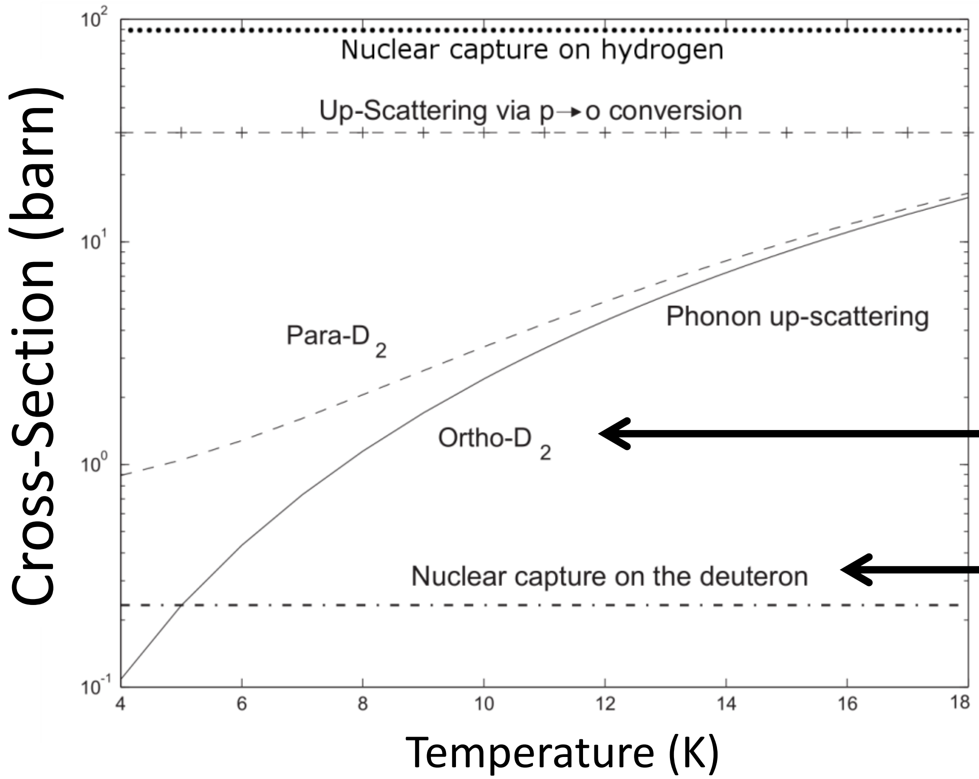


Neutron guides towards experiments

UCN flying upwards into storage volume

Loss of UCN in D_2

- Loss rate of UCN in D_2 : $\lambda = \lambda(process\ 1) + \lambda(process\ 2) + \cdots$
  → small λ desired
- $\lambda\ (process) = N_{scatterers} * \sigma_{process} * v_{UCN}$ → decrease $N_{scatterers}$ or $\sigma_{process}$



Suppress H and pD_2 content

D_2 has two spin isomers (like H_2) called ortho (oD_2) and para (pD_2) with $E_0(oD_2) < E_0(pD_2)$, $\Delta E = 7$ meV

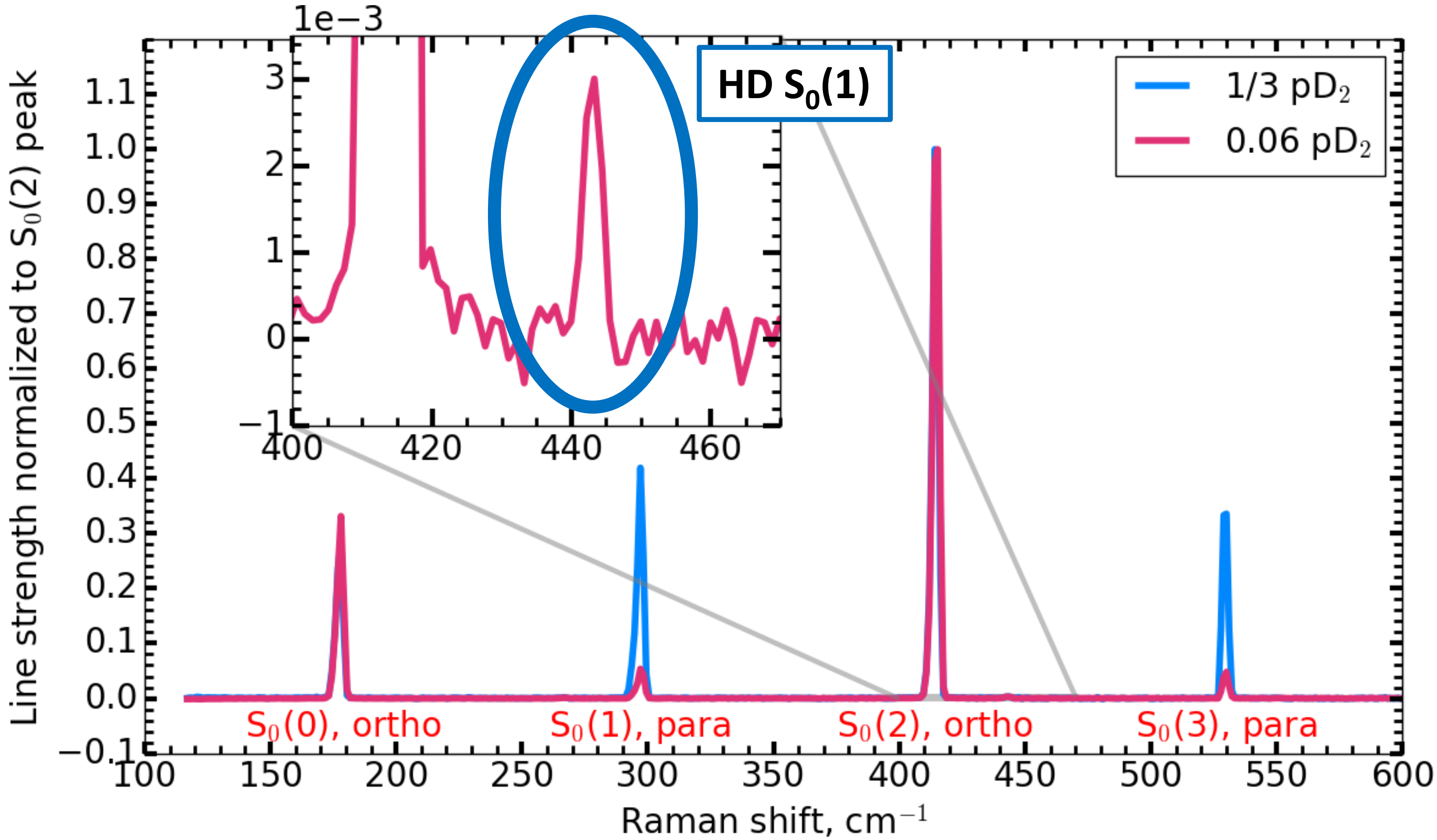
Keep temperature at 5 K

Limiting factor in a sD_2 moderator

Modified plot from Liu, C.-Y., Young, A. R., and Lamoreaux, S. K.: 2000, Phys. Rev. B 62, R3581

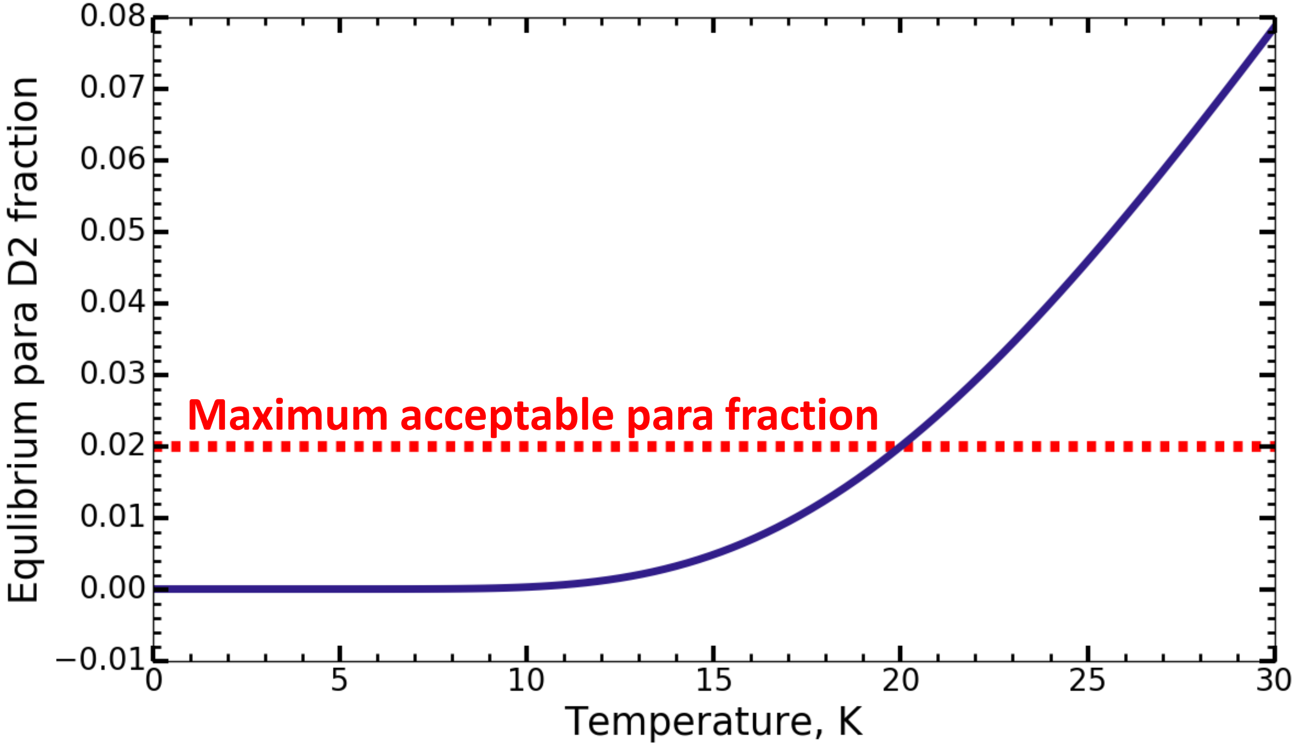
Monitoring Para-D_2 and HD Concentrations

- The **pD_2 and HD** concentrations are **determined using Raman spectroscopy**
- The concentrations are computed from the relative line strengths in a D_2 sample



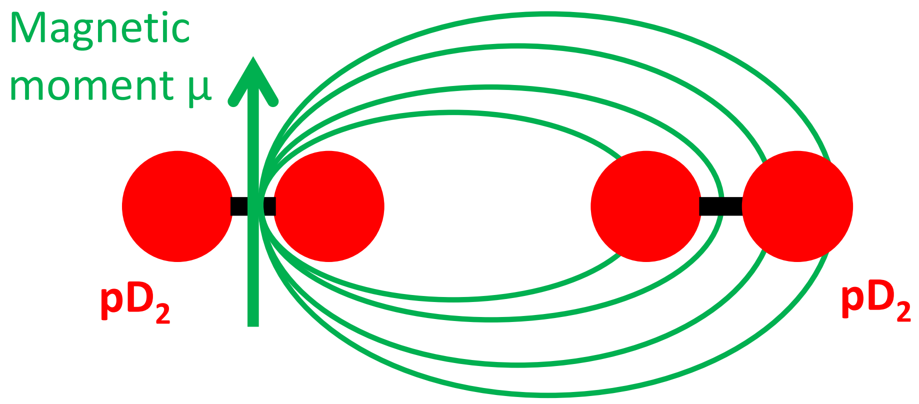
Decreasing the pD_2 Content

- **Maximum acceptable pD_2 fraction = 0.02, 0.005 to drop to nuclear absorption level**
- Thermal equilibrium ortho and para fractions follow Boltzmann statistics
- When freezing D_2 gas with a higher para fraction, this fraction decreases towards the equilibrium value of the new temperature → seems quite straightforward



Conversion Processes: Natural Self-Conversion

- Magnetic dipole-dipole and nuclear quadrupole **interactions between neighboring D_2 molecules can lead to spin realignment in pD_2 molecules, converting them to oD_2**
- Interaction is possible between 2 pD_2 or 1 oD_2 and 1 pD_2



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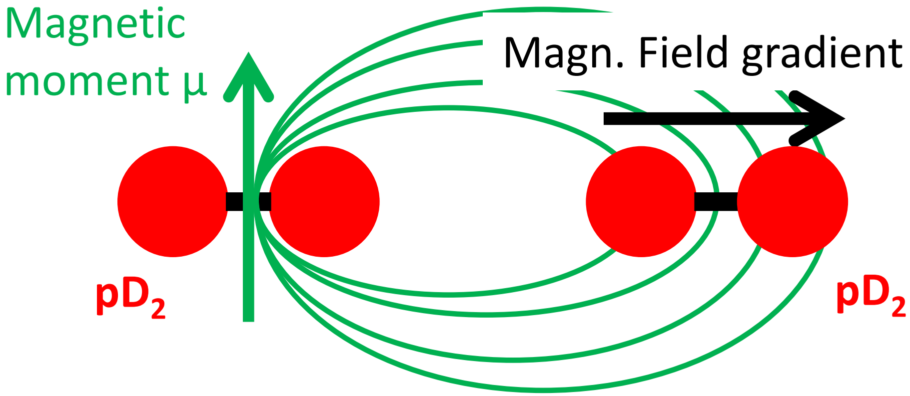

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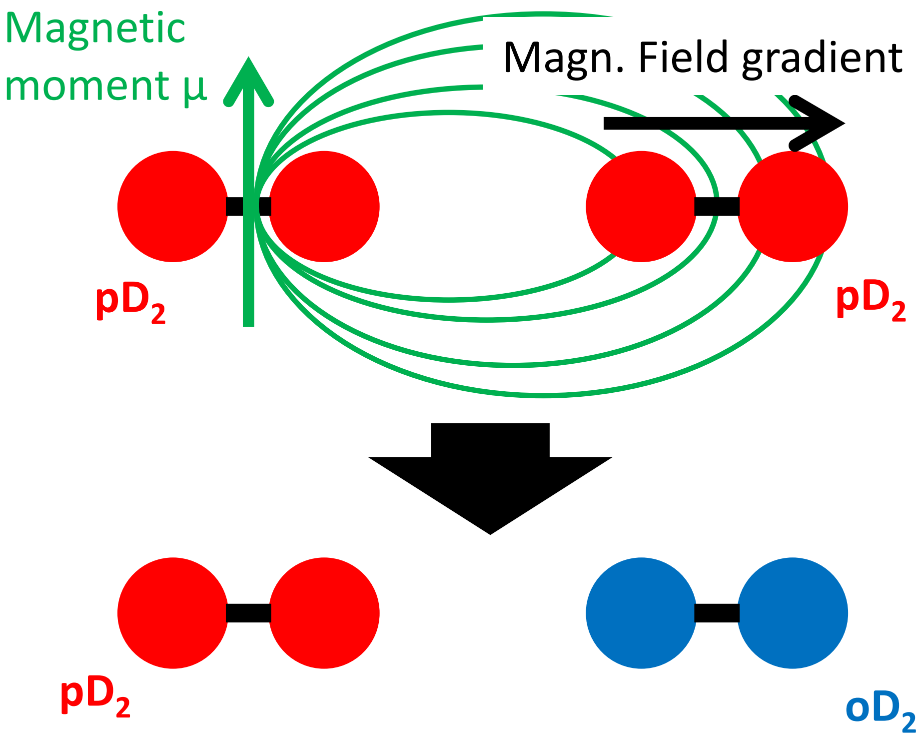

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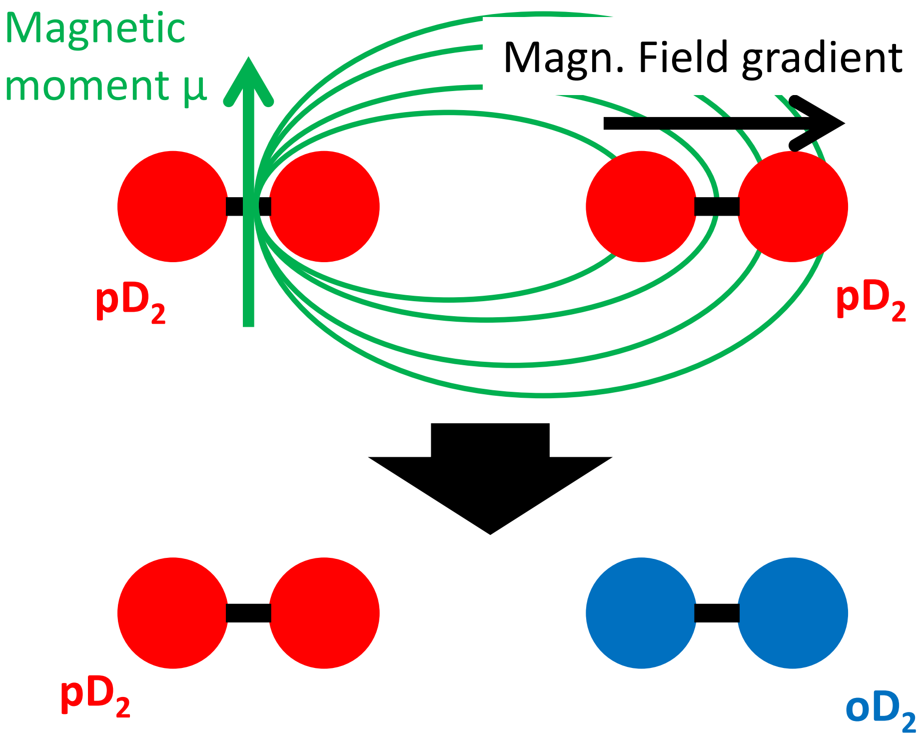
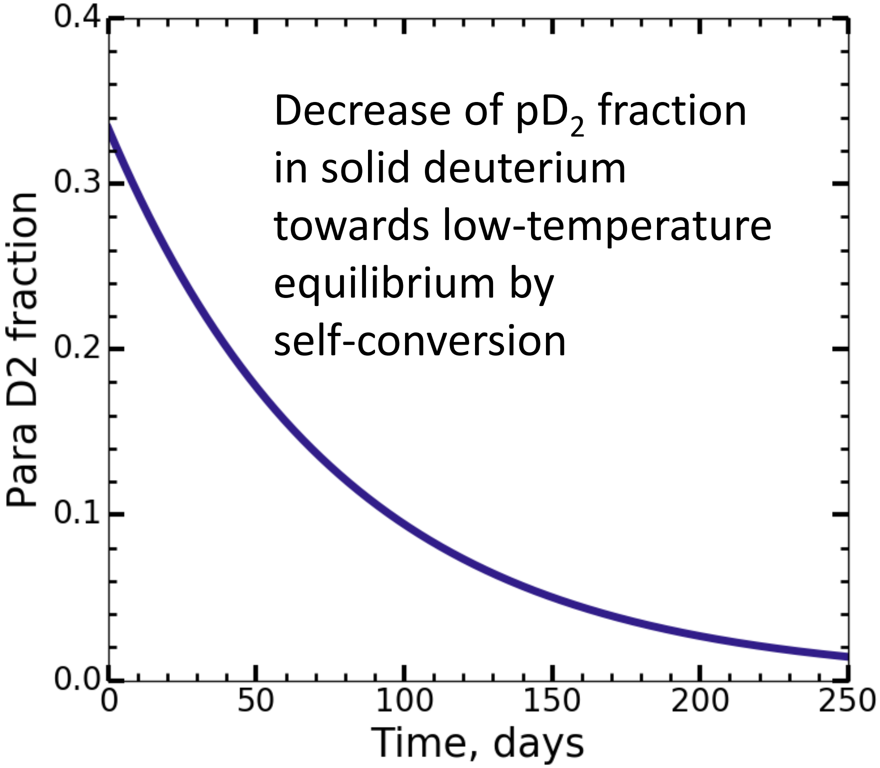
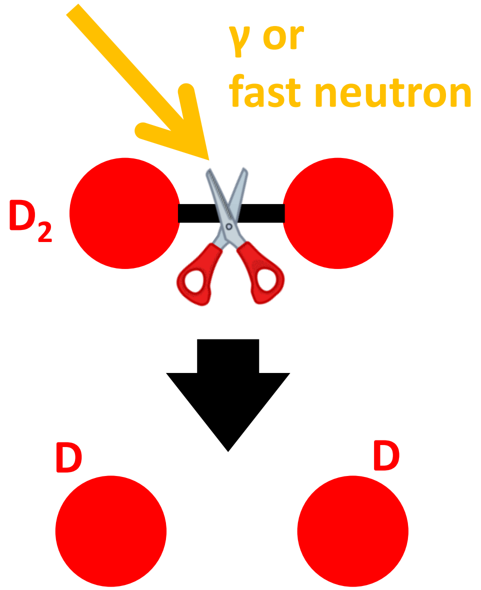
- **Slow process**: τ = 1900 h in 4 K solid D_2 → **7 months from 0.33 to 0.02 para fraction!**





Conversion Processes: Catalysis by Irradiation

- **Fraction of D_2 molecules gets broken up** into single deuteron atoms **by fast neutrons and γ radiation** from the spallation target



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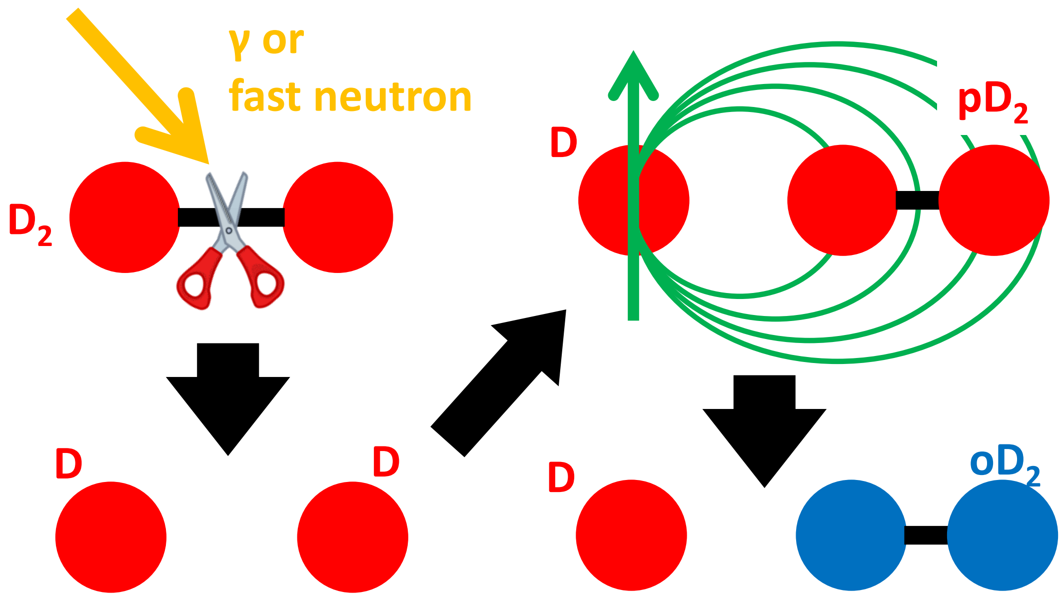
- **Fraction of D_2 molecules gets broken up** into single deuteron atoms **by fast neutrons and γ radiation** from the spallation target
- **D atoms** diffuse through the D_2 crystal and **generate large magnetic field gradients, which can convert para molecules** along the deuteron's path



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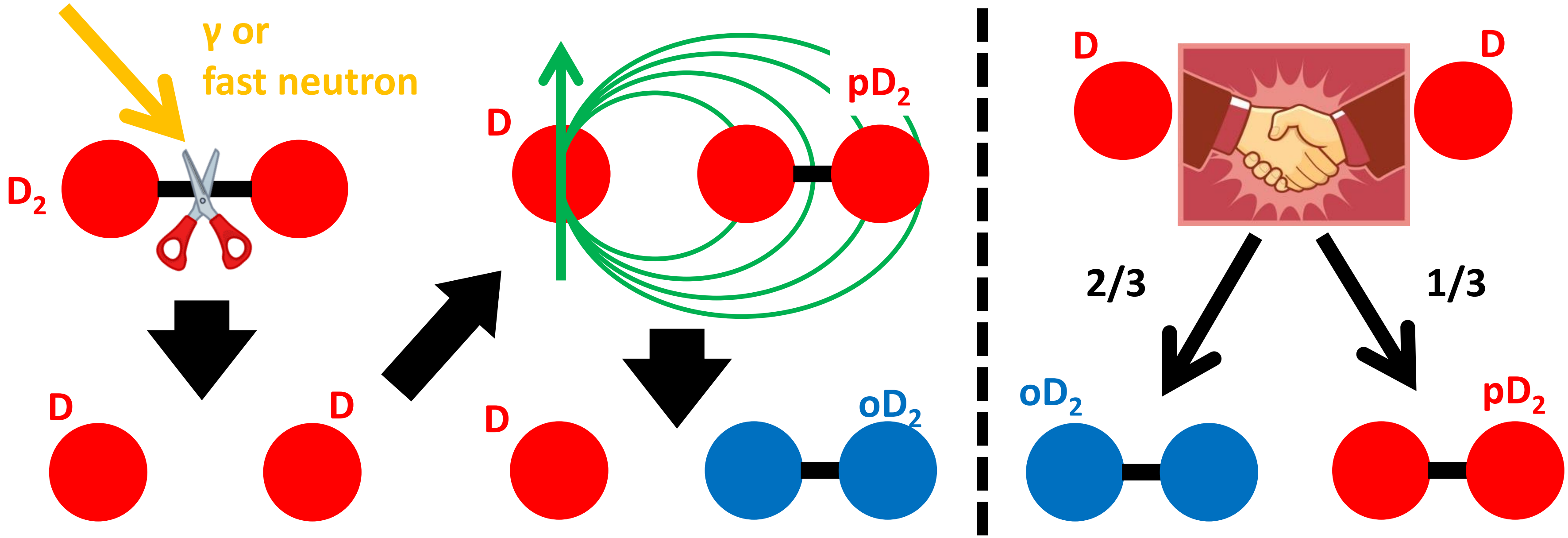
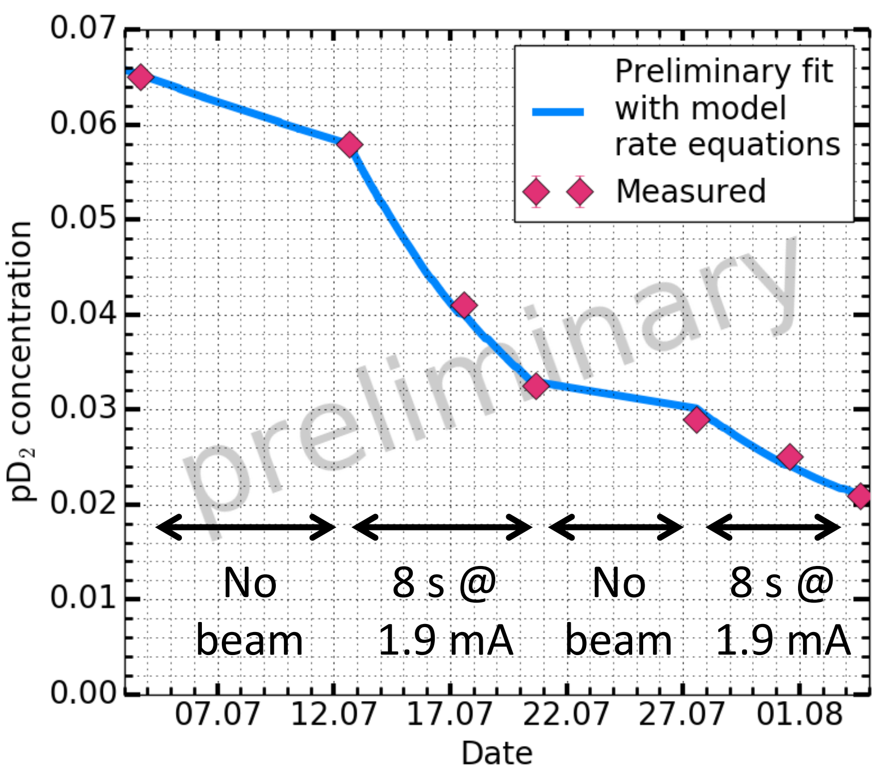
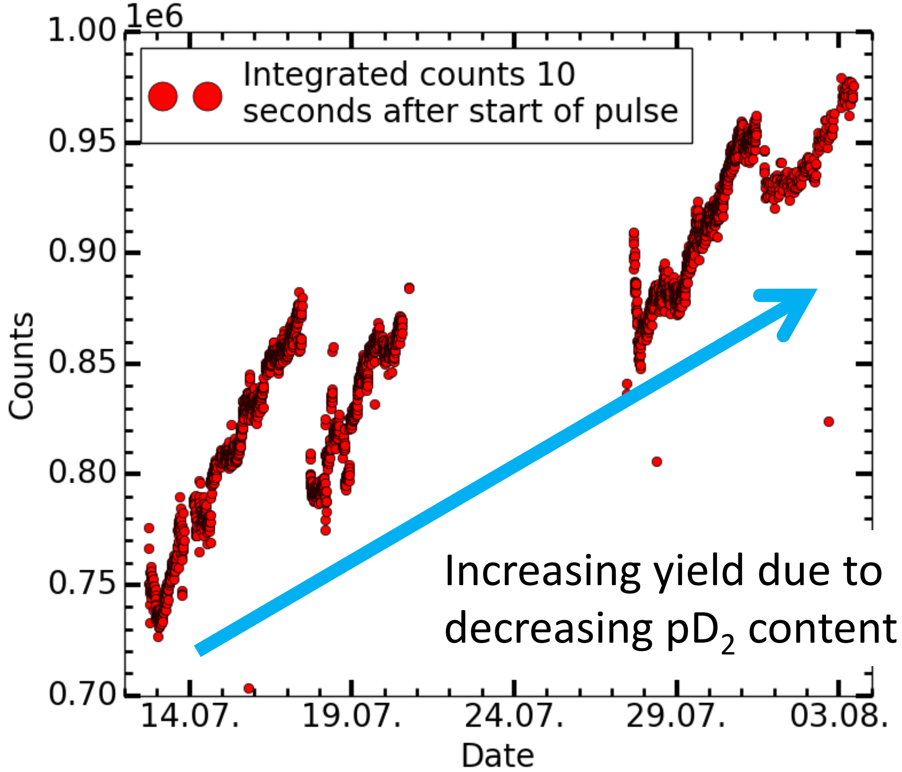
- **D atoms** diffuse through the D_2 crystal and **generate large magnetic field gradients, which can convert para molecules** along the deuteron's path
- **Competing process: recombination of deuterons produces room temperature equilibrium D_2** i.e. 2/3 oD_2 and 1/3 pD_2



Evolution of pD_2 Content in the PSI UCN Source Moderator

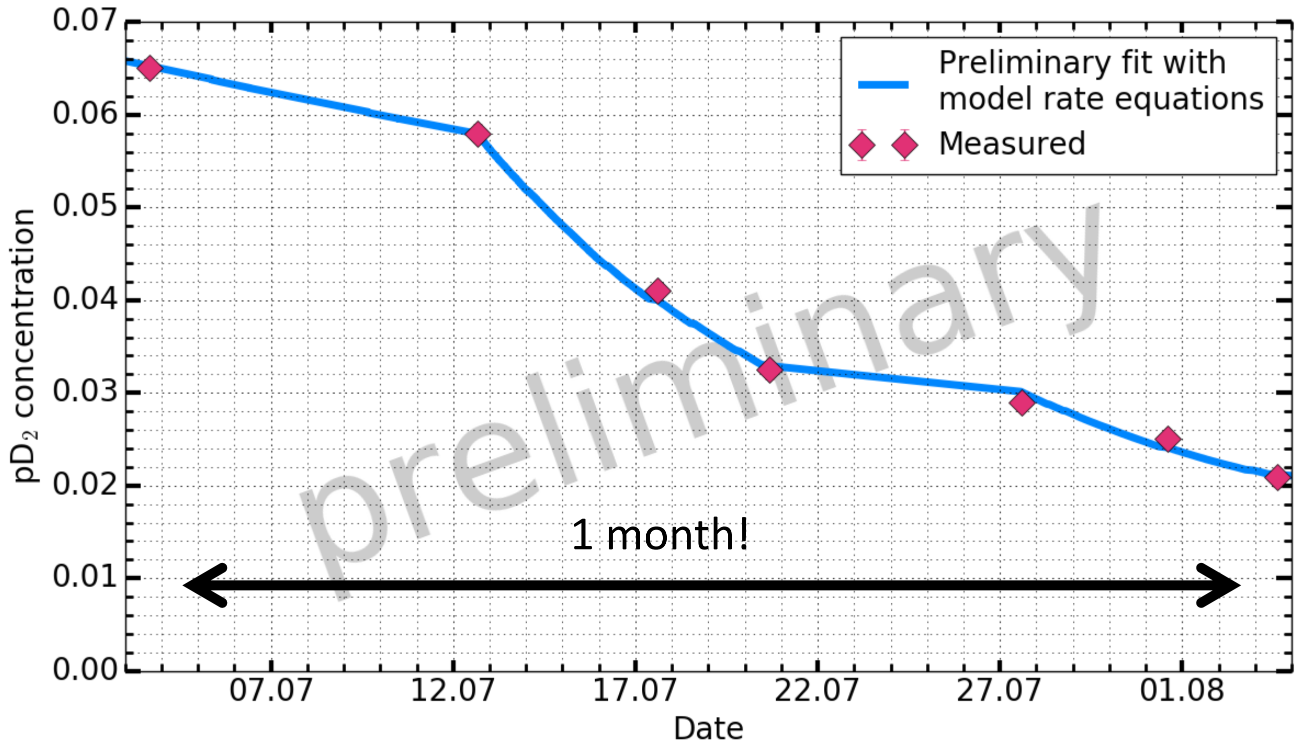
- **680 g D_2 with initial 0.075 para fraction frozen in moderator, monitoring of evolution of pD_2 content by self-conversion and during pulsing** (8 s pulses every 300 s)
- **Data analysis ongoing, points of interest** include **limits on power deposition** in D_2 and **changes in macroscopic D_2 structure** during operation





Evolution of pD_2 Content in the PSI UCN Source Moderator

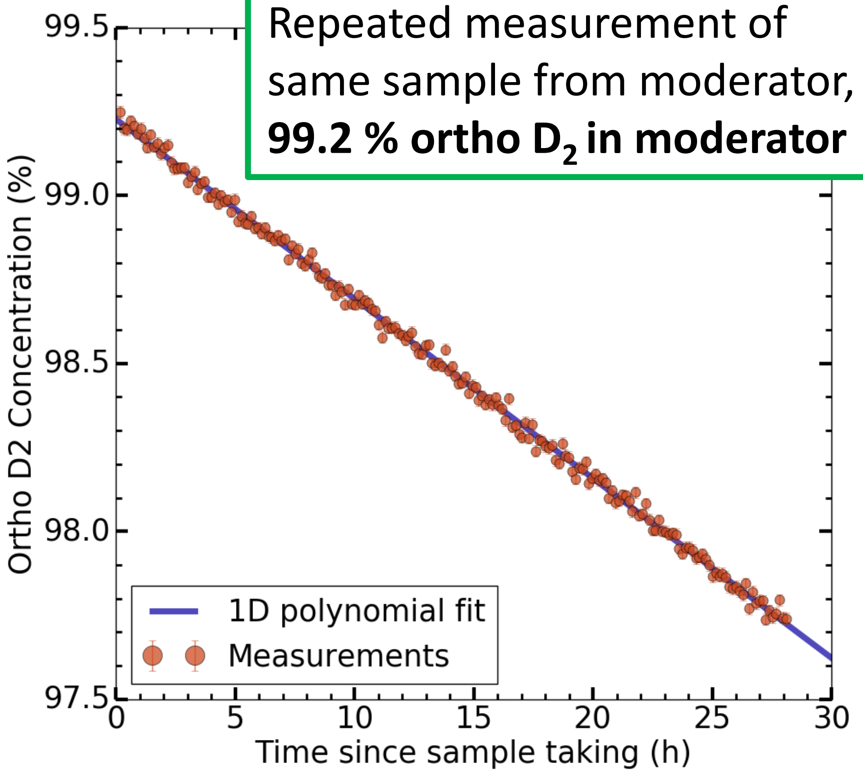
- Clear that **conversion towards acceptable pD_2 fraction just by self-conversion and pulsing is too slow**
- Need another method to ensure high oD_2 concentration even before D_2 is transferred to the moderator vessel



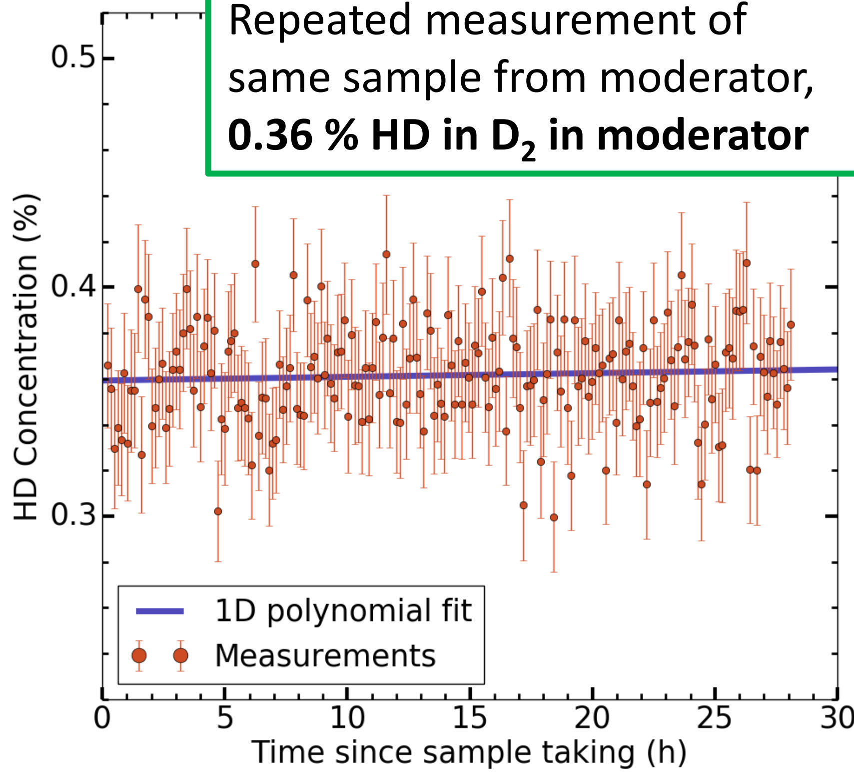
Monitoring pD_2 and HD Concentrations

- **Accelerate conversion in liquid D_2** at about 20 K before transfer **using Oxisorb®**
- **Starting concentration of 98 % oD_2**
- Both oD_2 and HD concentrations **within acceptable limits from the start**

Repeated measurement of same sample from moderator, **99.2 % ortho D_2 in moderator**

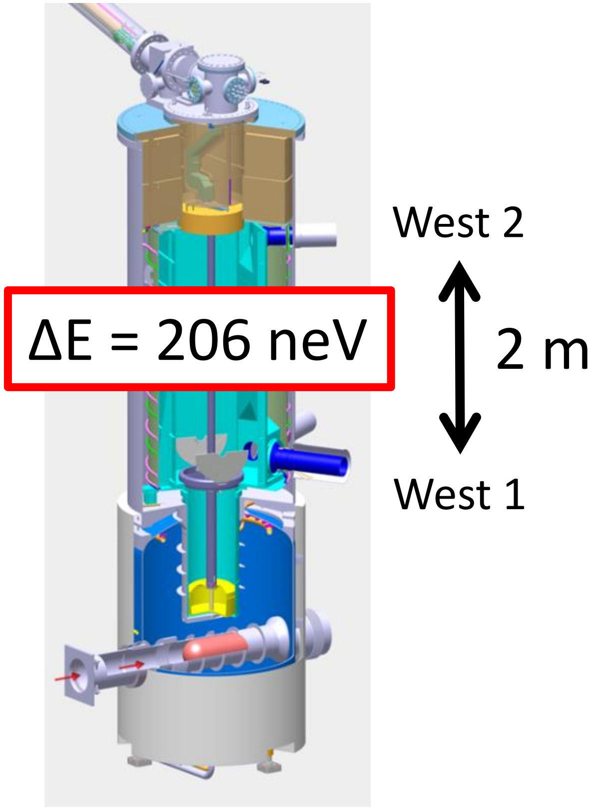


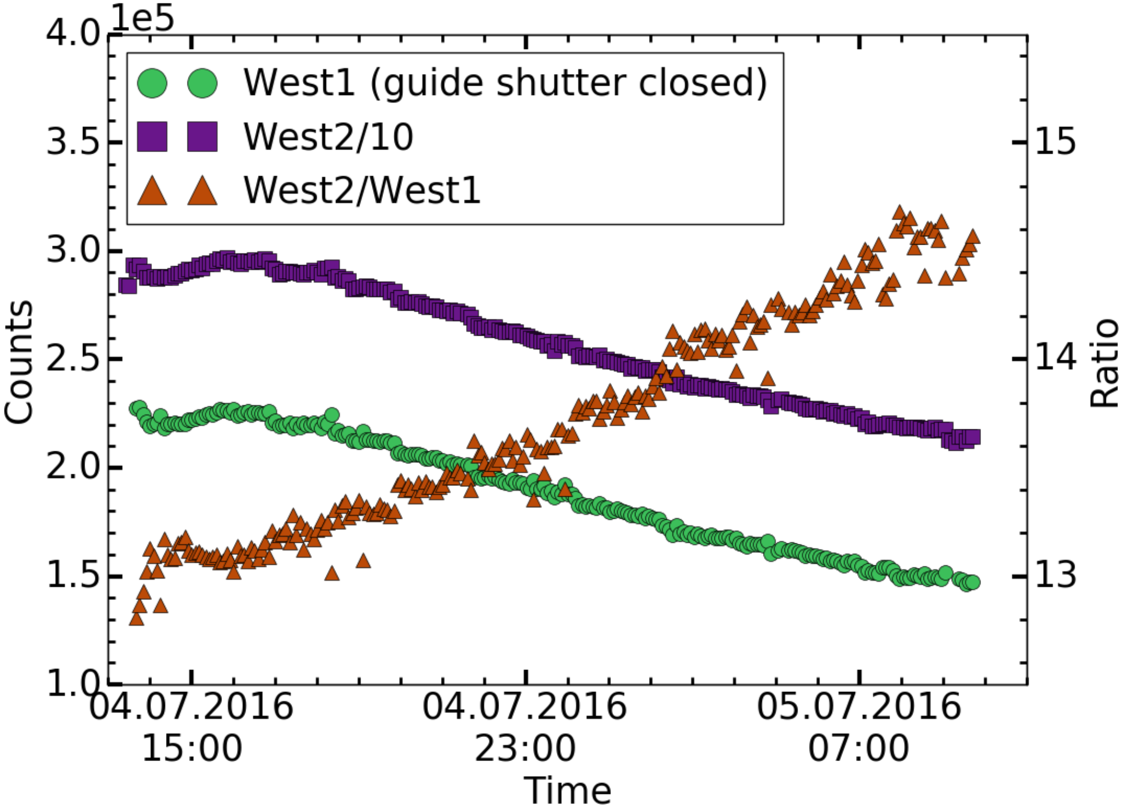
Repeated measurement of same sample from moderator, **0.36 % HD in D_2 in moderator**



Short-Term UCN Yield Decrease

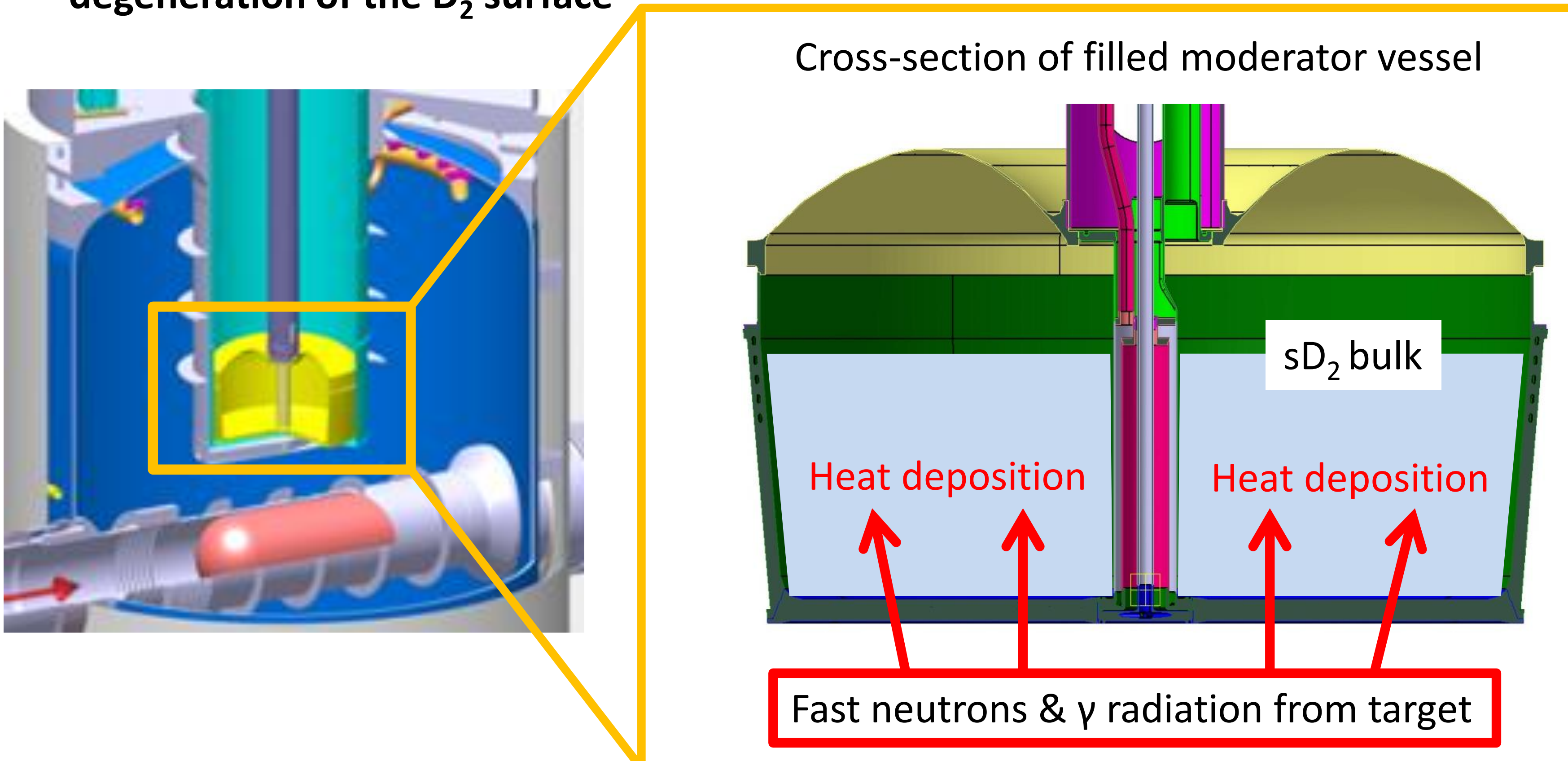
- **Continuous pulsed operation leads to a decrease of UCN output**
- UCN count ratio West2/West1 increases → **UCN intensity decreases more rapidly for slower UCN**





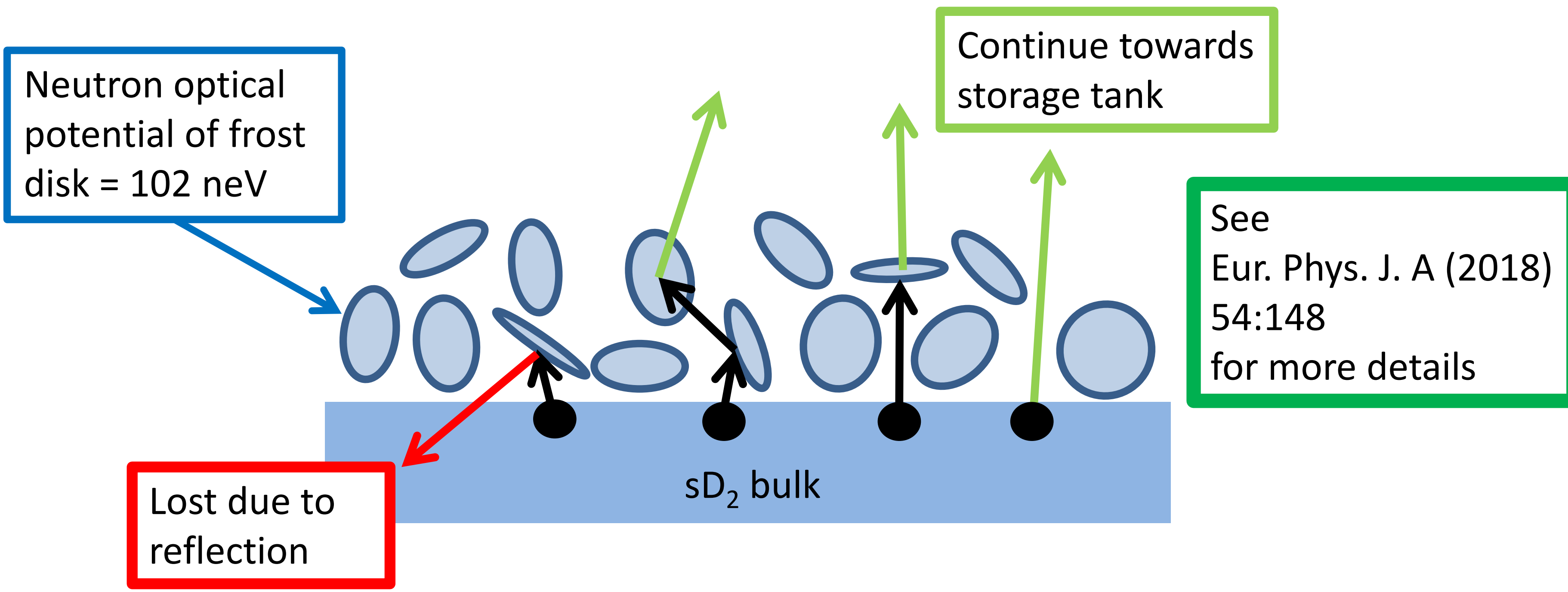
Frost Model

- Most attractive idea to **explain the energy-dependent decrease: frost hypothesis**
- **Heat deposition in D_2 due to fast neutrons and γ radiation during pulse leads to degeneration of the D_2 surface**



Frost Model

- Simplified picture : layers of **small D_2 frost disks with neutron optical potential of 102 neV** form **on the bulk surface** that **increase the scatter of exiting neutrons**
- Continuous pulsing → number of layers ↗ → transmission probability ↘
- **UCN with E_{kin} < 102 neV totally reflected, for E_{kin} > 102 neV the reflection probability decreases with increasing E_{kin}**

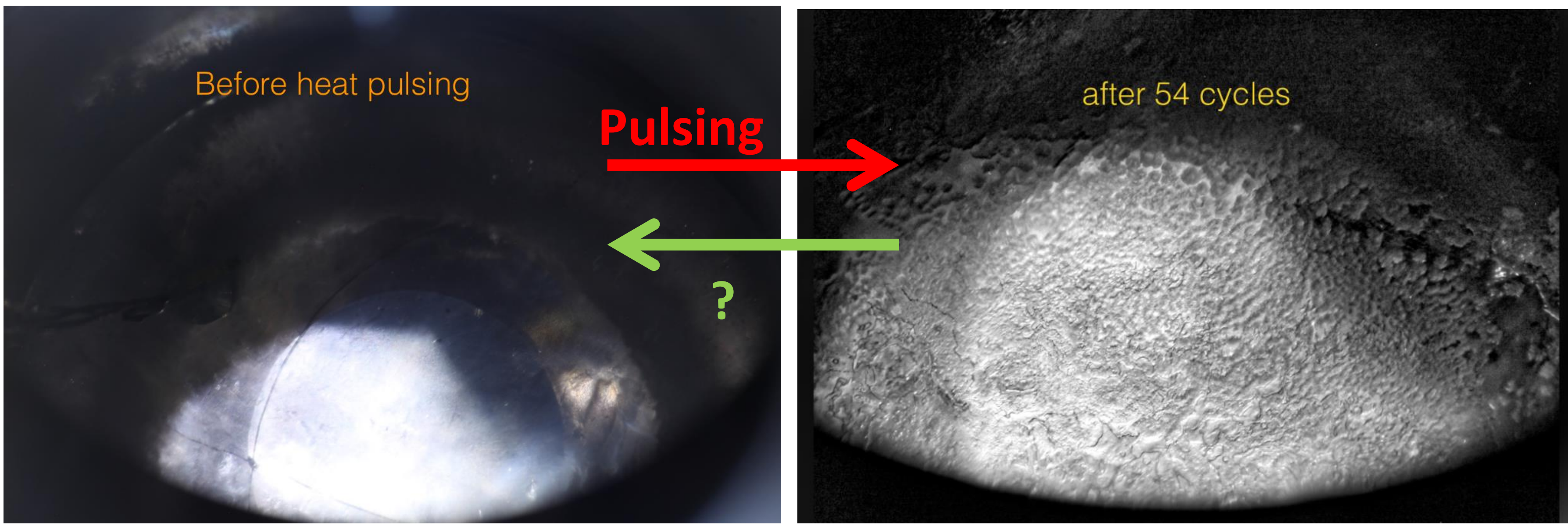


Optical observation of sD_2 at UCN source at PULSTAR reactor

- Visual confirmation of solid D_2 surface degradation after heat cycling, possible because setup not yet inserted into reactor contrary to PSI source moderator vessel
- **Need a procedure to reverse surface degradation**

Transparent D_2 crystal with smooth surface

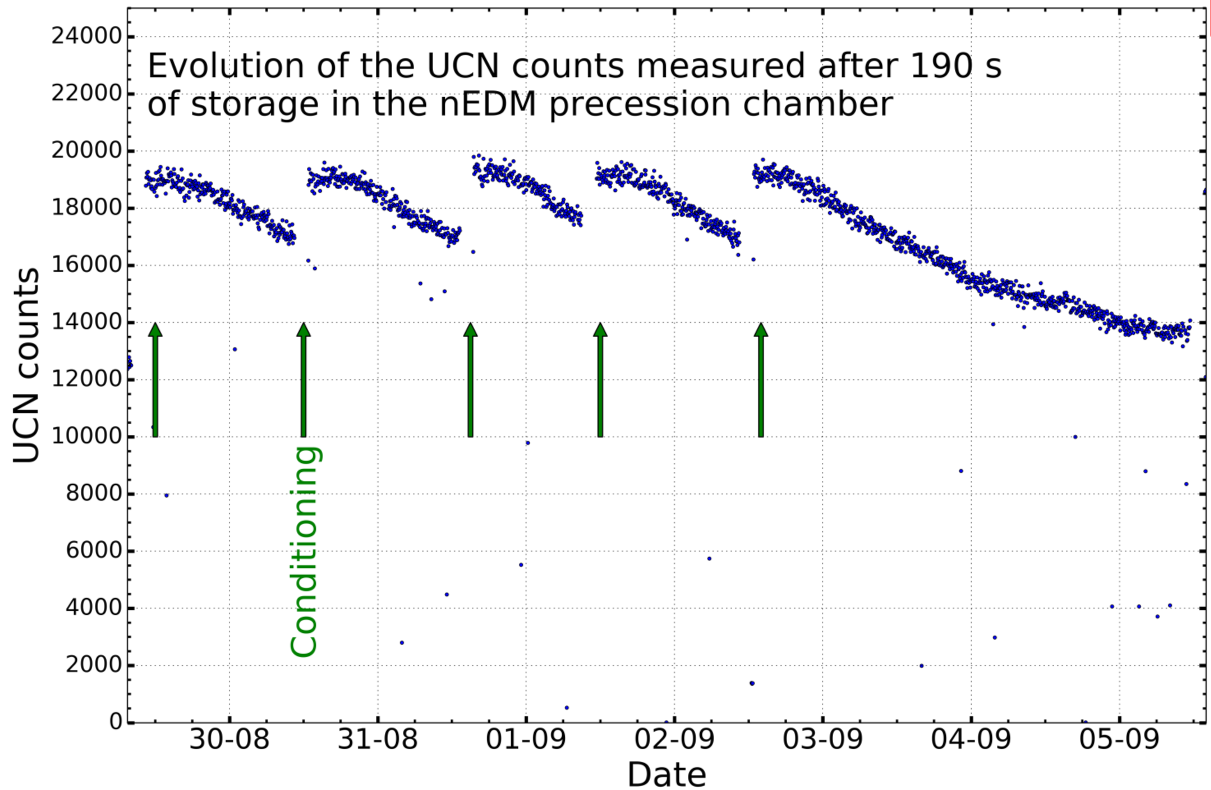
Rough surface after several heat cycles

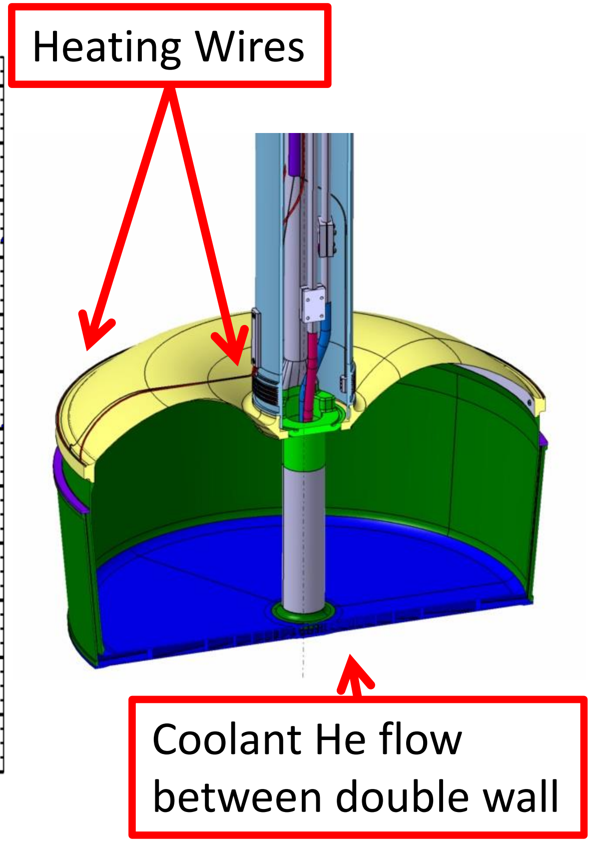


Photos by E. Korobkina and group, NC State University

Conditioning

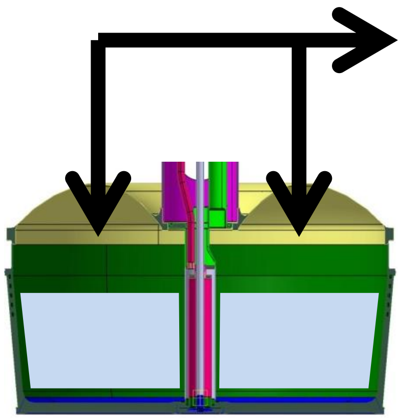
- Development of **surface treatment called "conditioning"** to recover output:
  Reduce He cooling of moderator vessel with additional heat input using heating wires
- But **conditioning interrupts operation** → **Minimize time** needed for output recovery

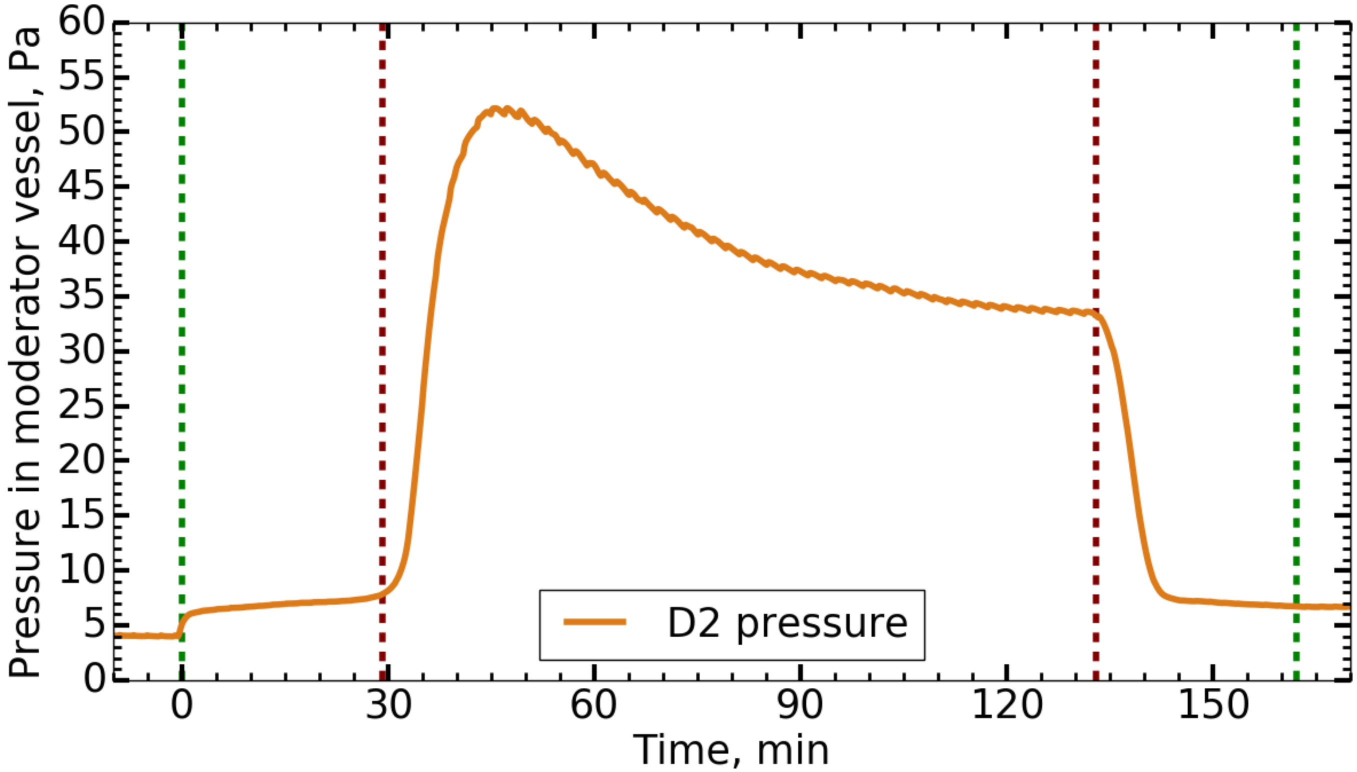




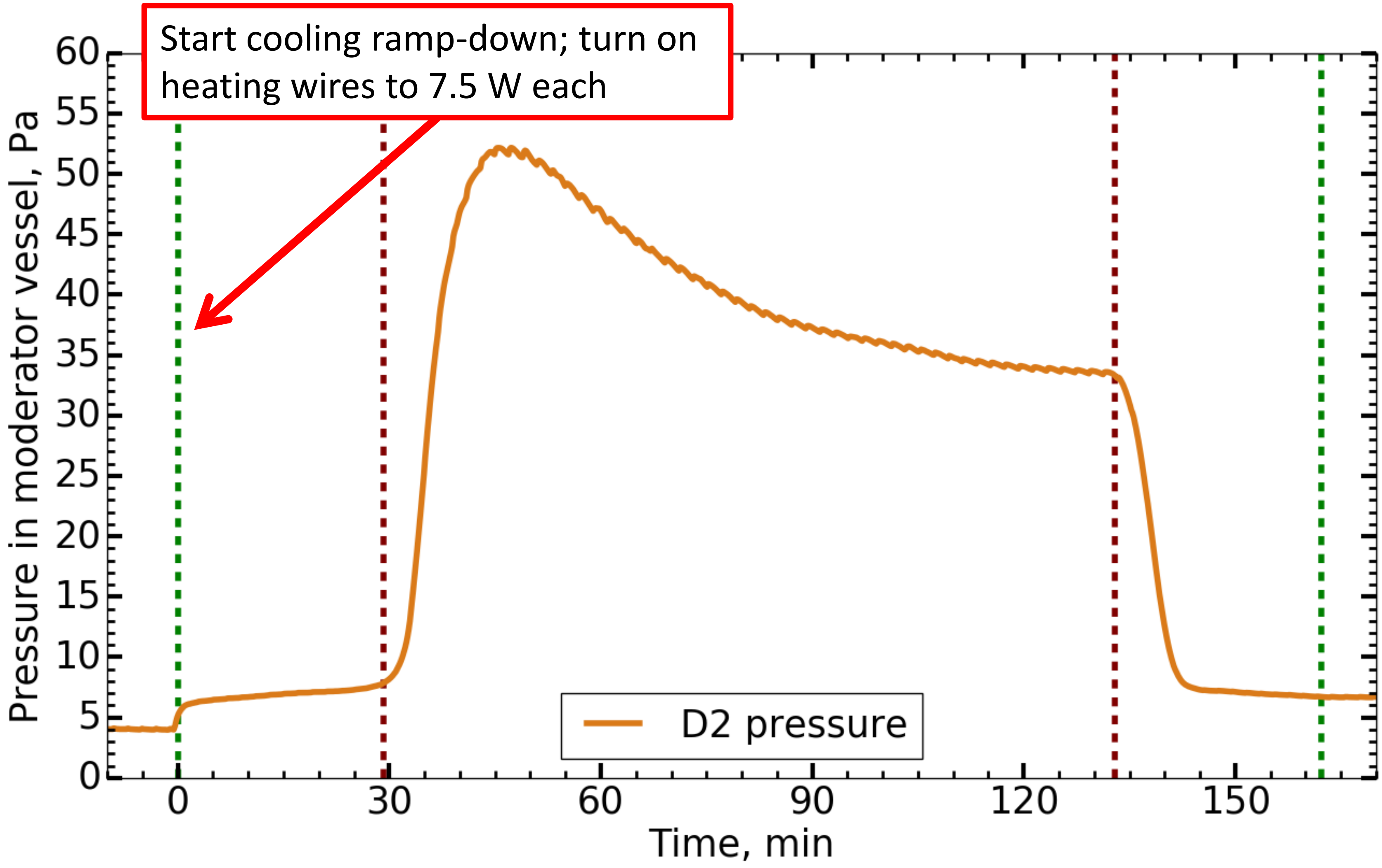
Conditioning Procedure

- Remember: **no visual inspection possible because source is a closed system**
- **Indirect observation of D_2 during conditioning through its vapor pressure** in moderator vessel





Conditioning Procedure



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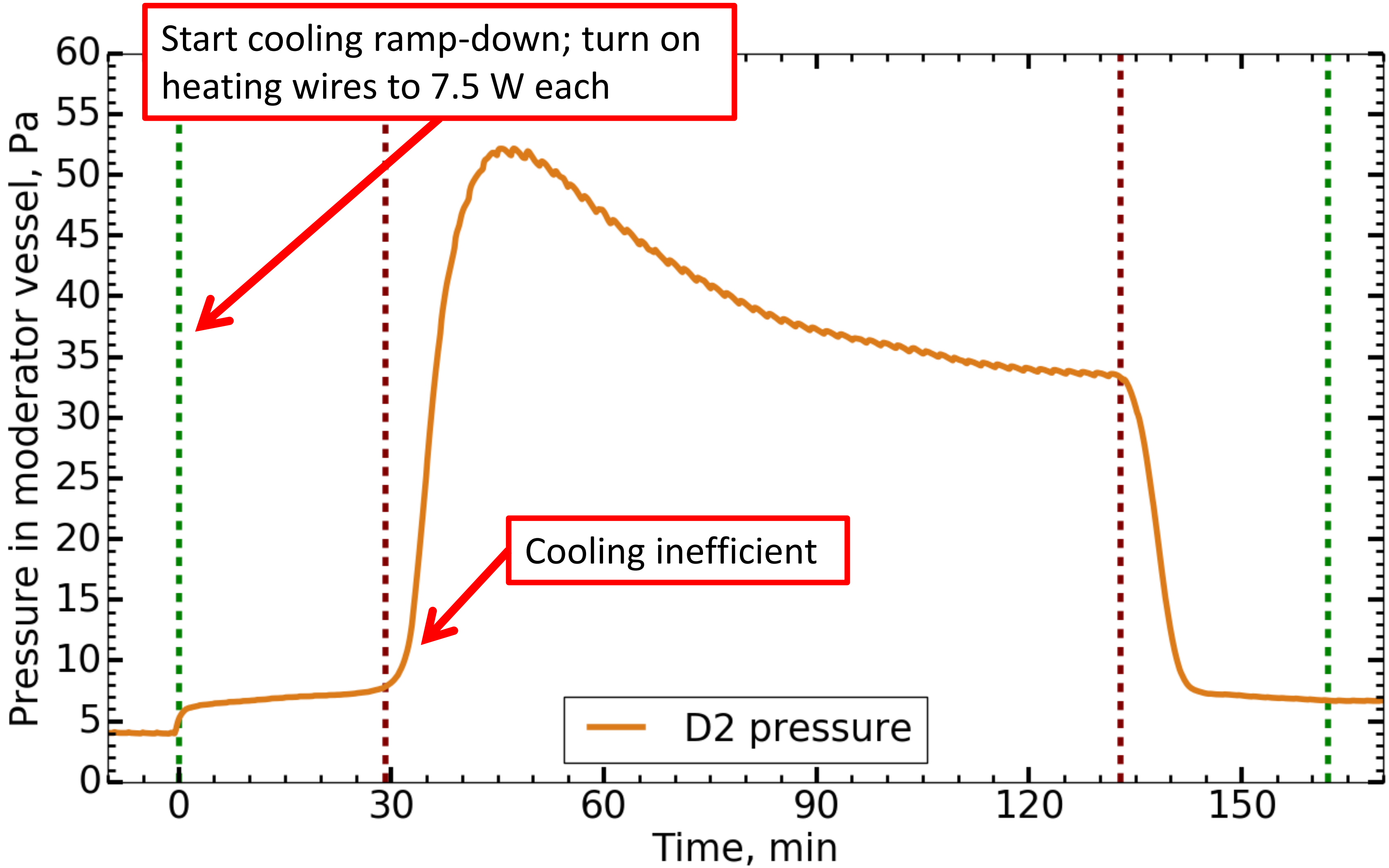

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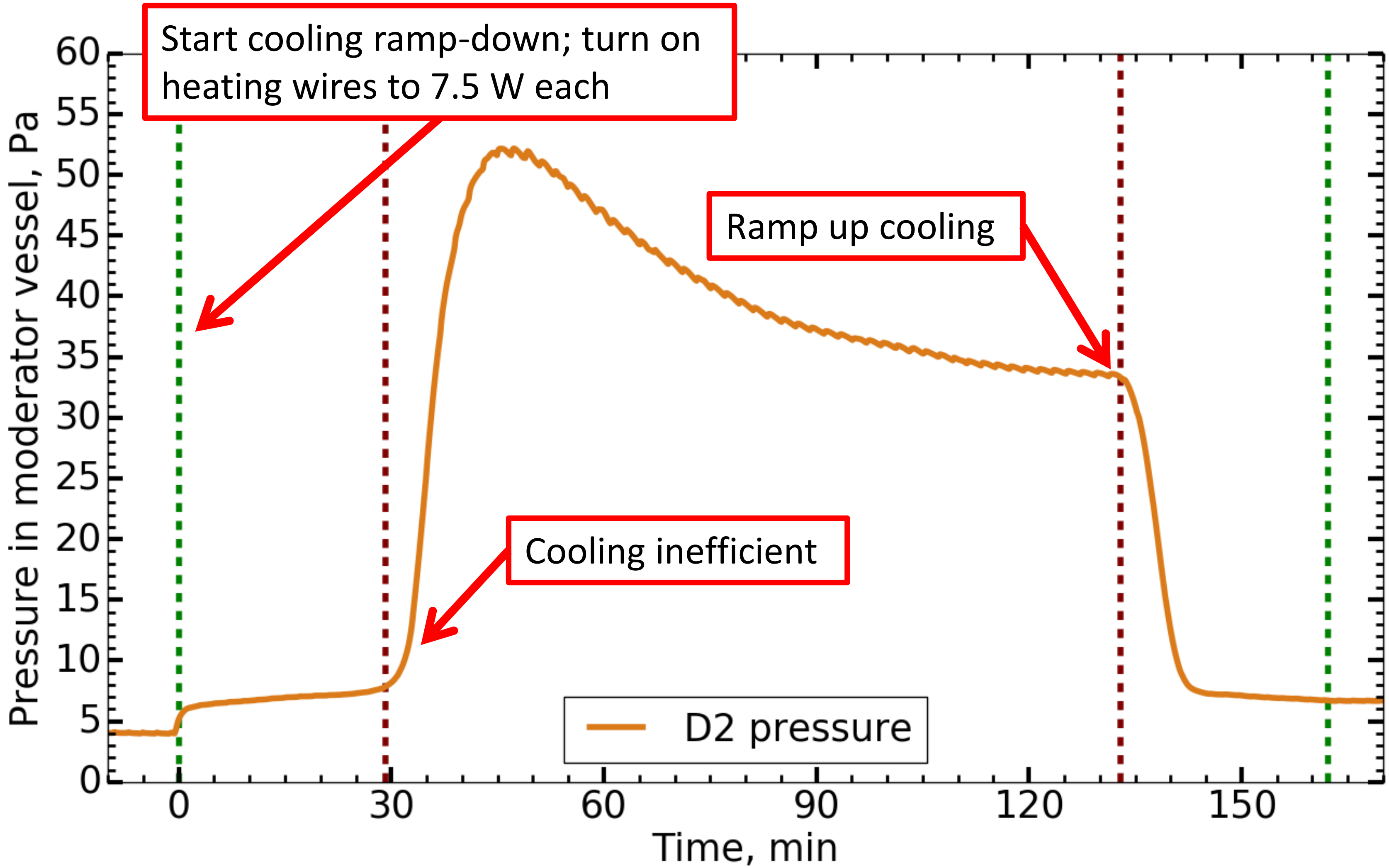

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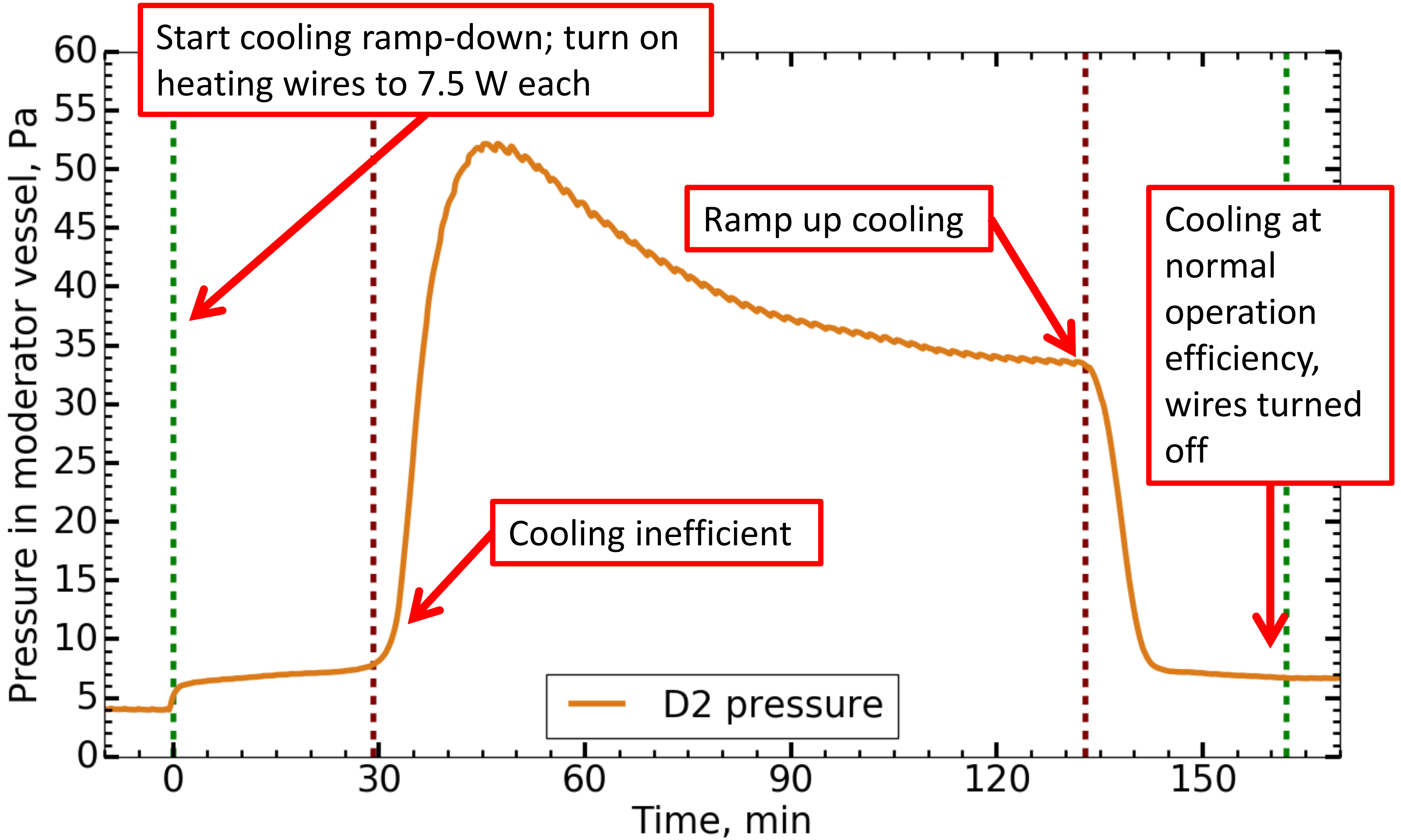
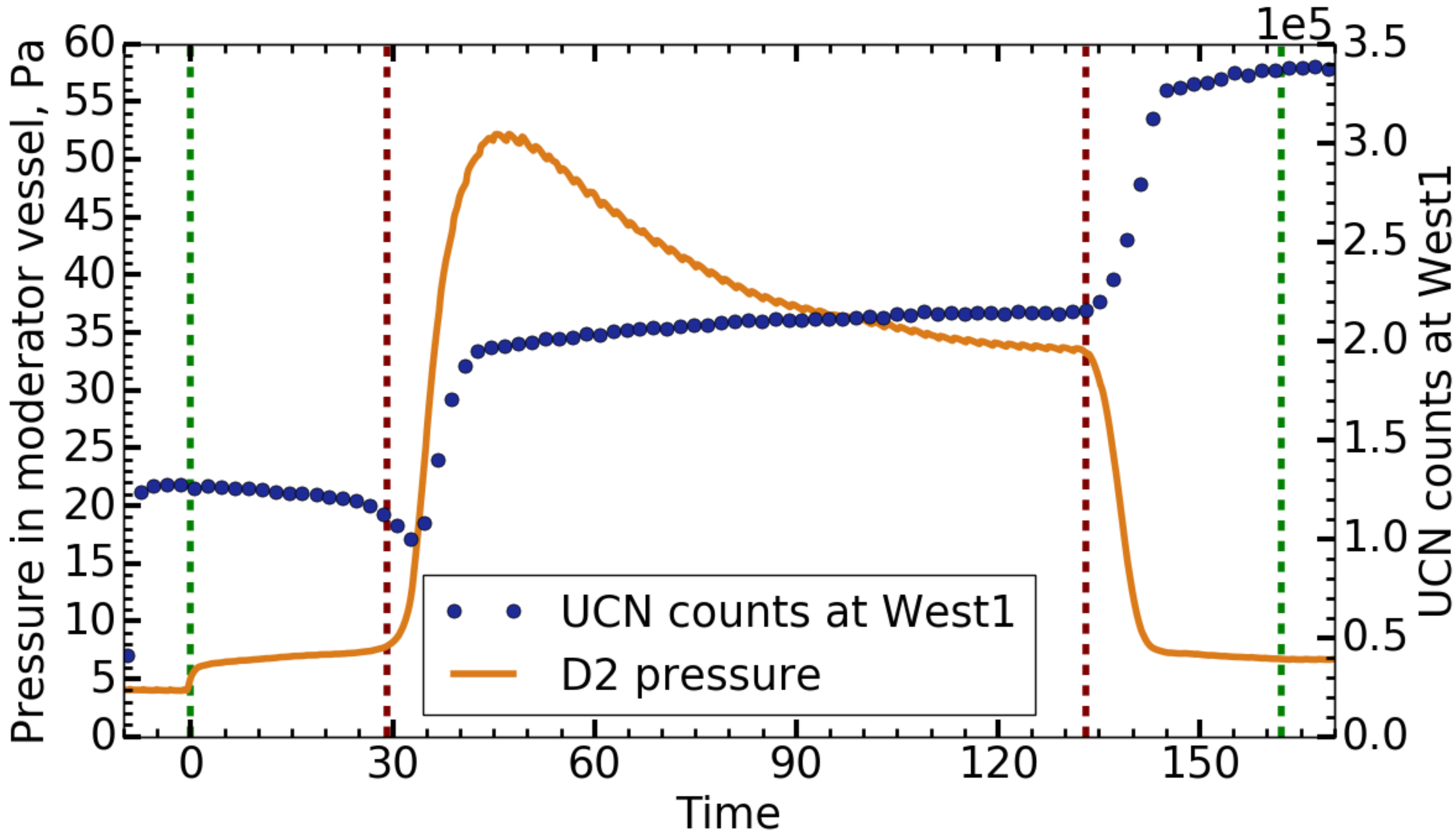
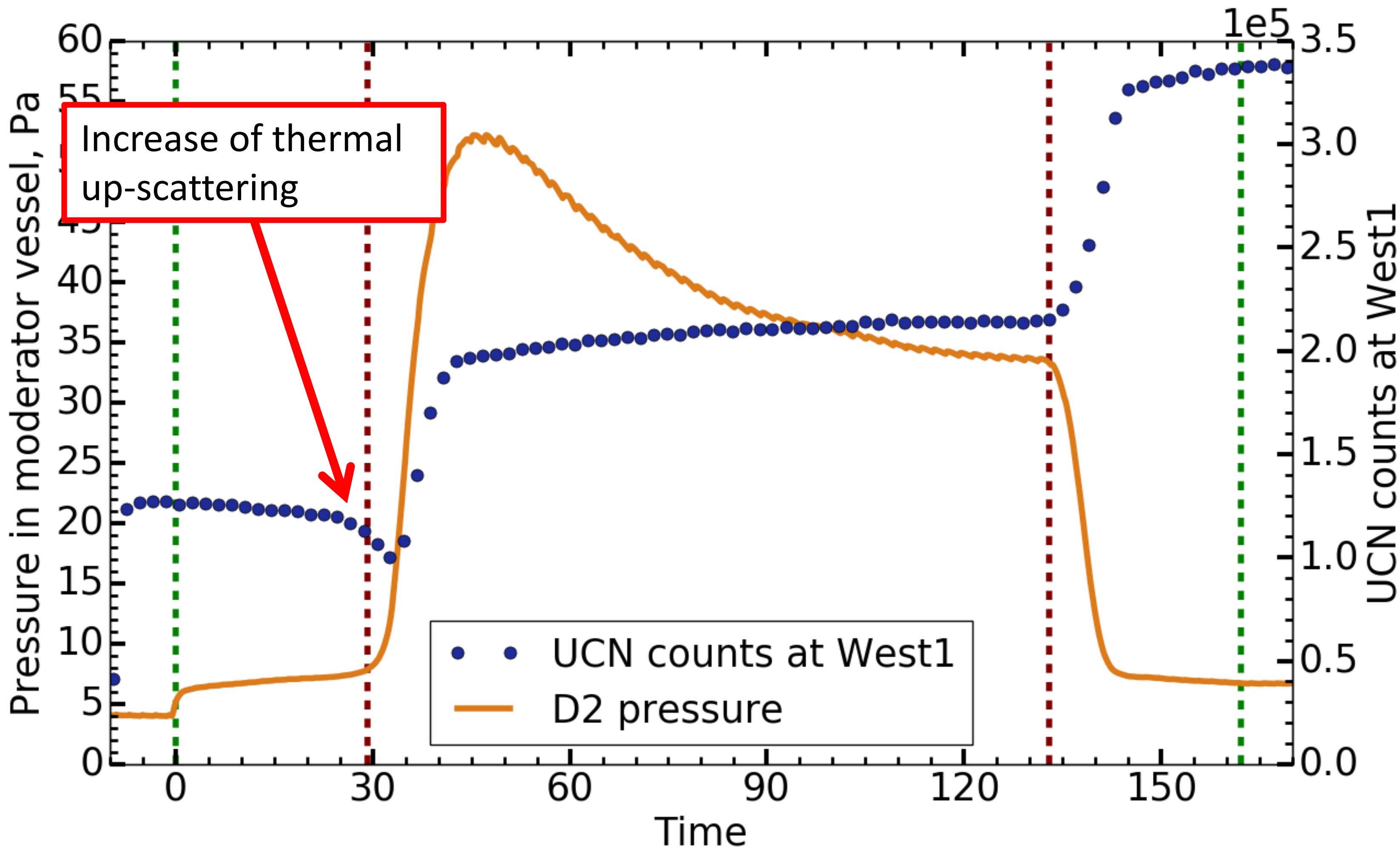


Probing UCN Output

- **Probe UCN output during conditioning** with short 0.1 s pulses at 1.4 mA in quick 2 min succession → minimal interference with conditioning process



Interpretation



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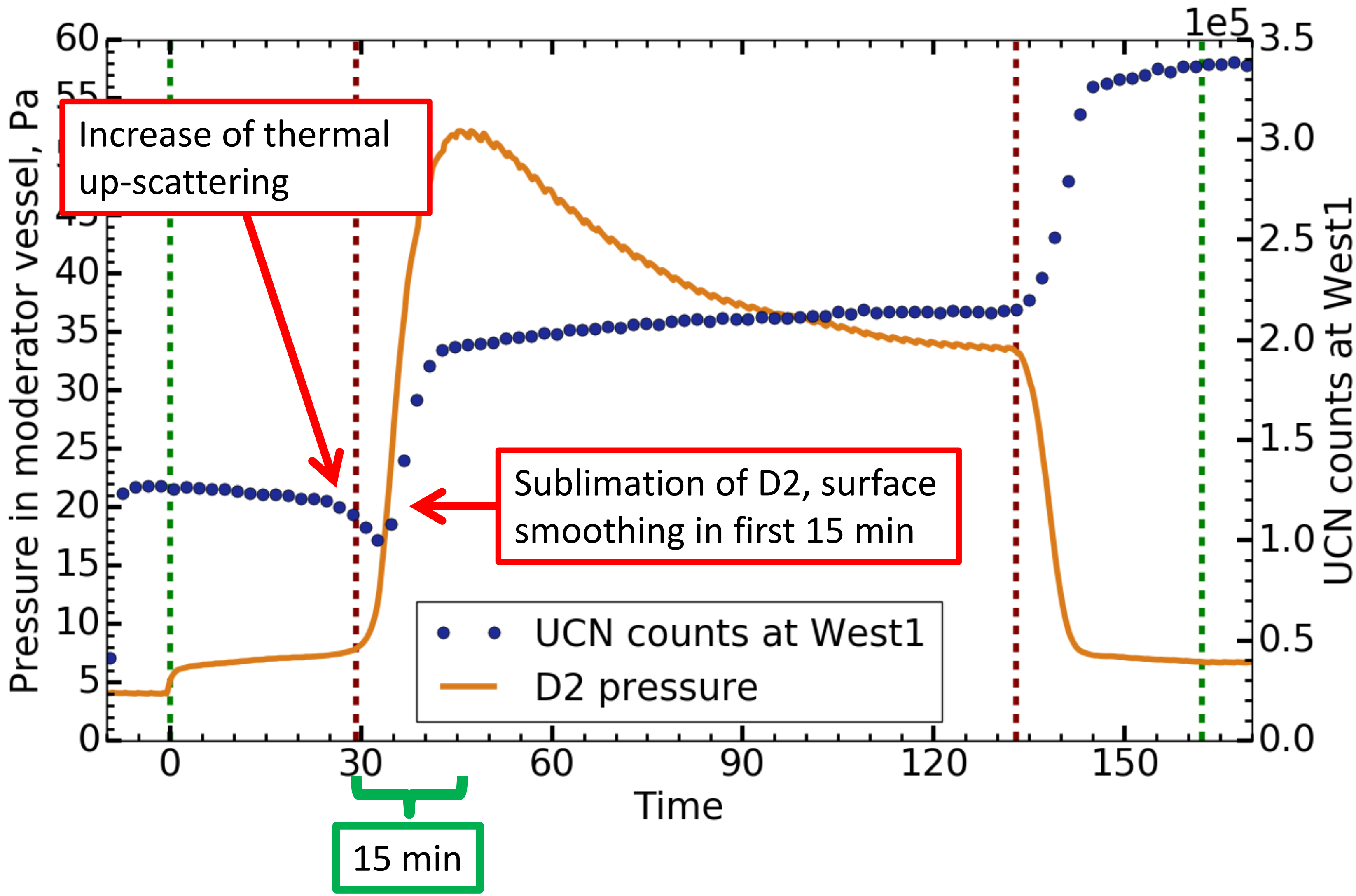

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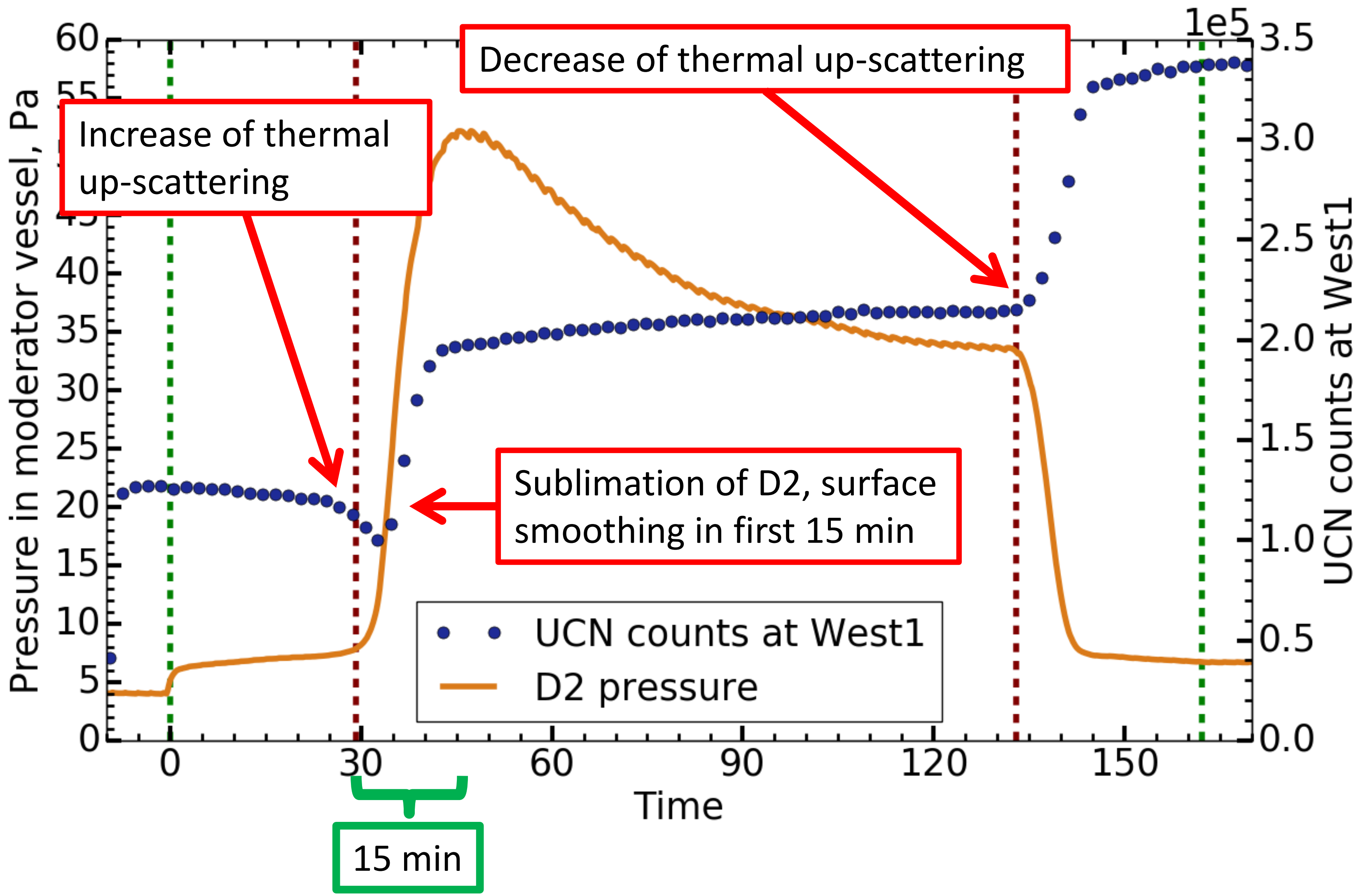
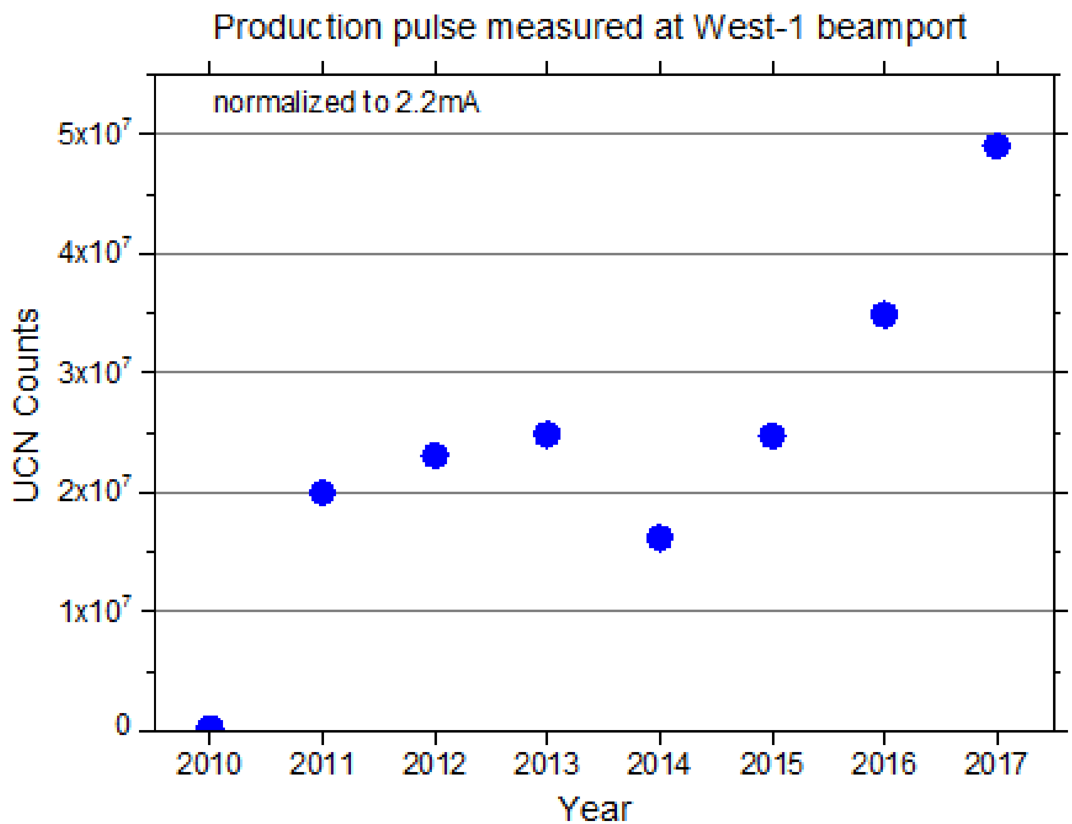


Resulting Output

- **Conditioning helped to reach new record UCN outputs at PSI**
- Insights will be used to **ensure high UCN output for next experiments and improve the sensitivity of the neutron EDM measurements**



Summary

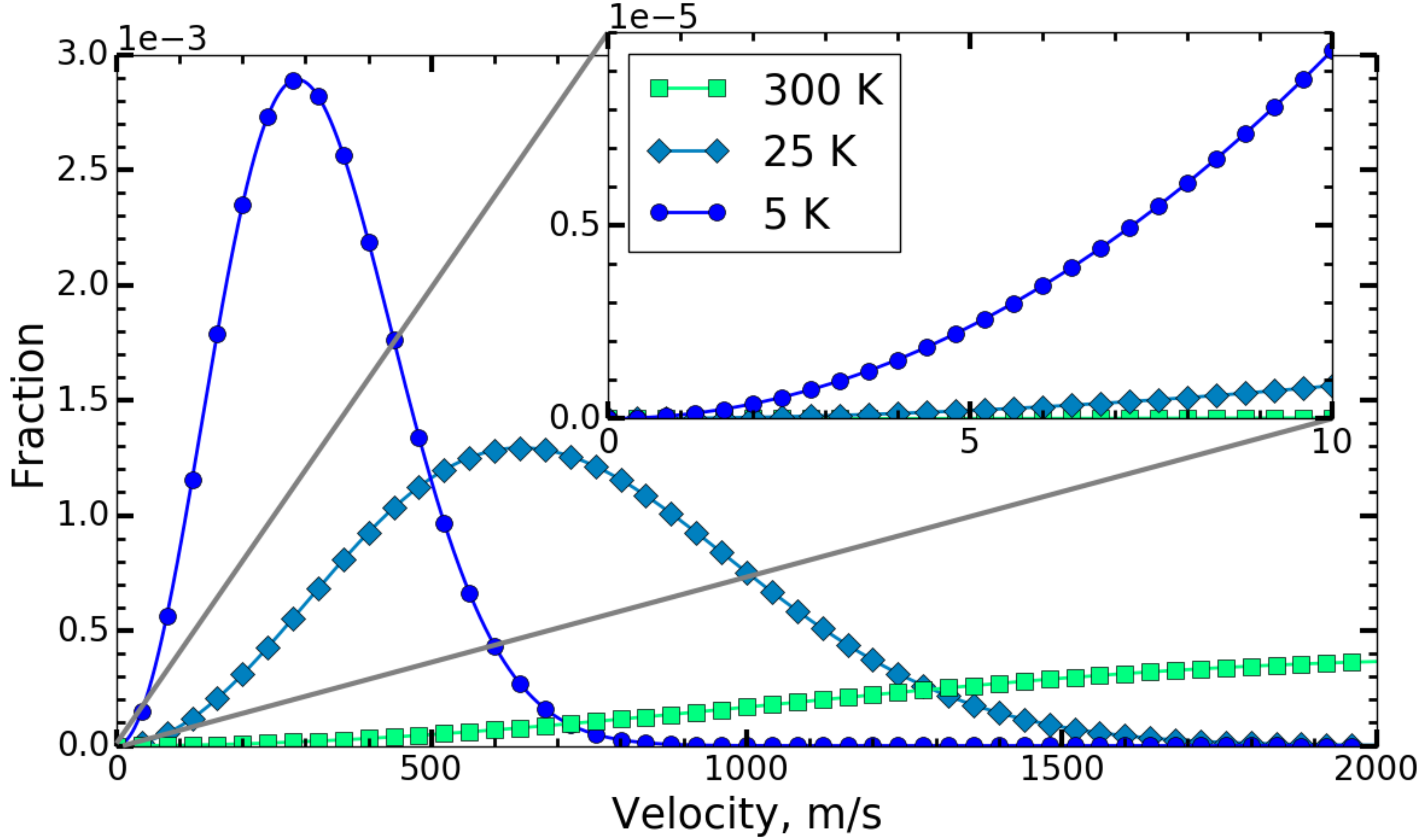
- **Accelerated para to ortho D_2 conversion due to irradiation has been measured for D_2 filled into the PSI UCN source moderator**, which may for example allow to determine limits for the energy deposition in D_2
- The **PSI UCN source shows short-term decrease** in its output, even though molecular losses are kept under control and impurities are monitored with Raman spectroscopy
- The short-term decrease can be explained with **D_2 frost forming on top of the bulk**
- **Conditionings**, short periods of reduced cooling and heat input, are applied to **counter the daily decrease**
- **Refinement of the conditioning** procedure has allowed the PSI source to **improve its average UCN output**, helping the **nEDM and future experiments** to further **improve their sensitivity**

Thank you for your attention

Backup Slides

Production of UCN

- **Extract the low-energy tail of a distribution of neutrons in thermal equilibrium with a medium (moderator)**, e.g. D_2O close to a nuclear reactor



Ortho/Para Deuterium

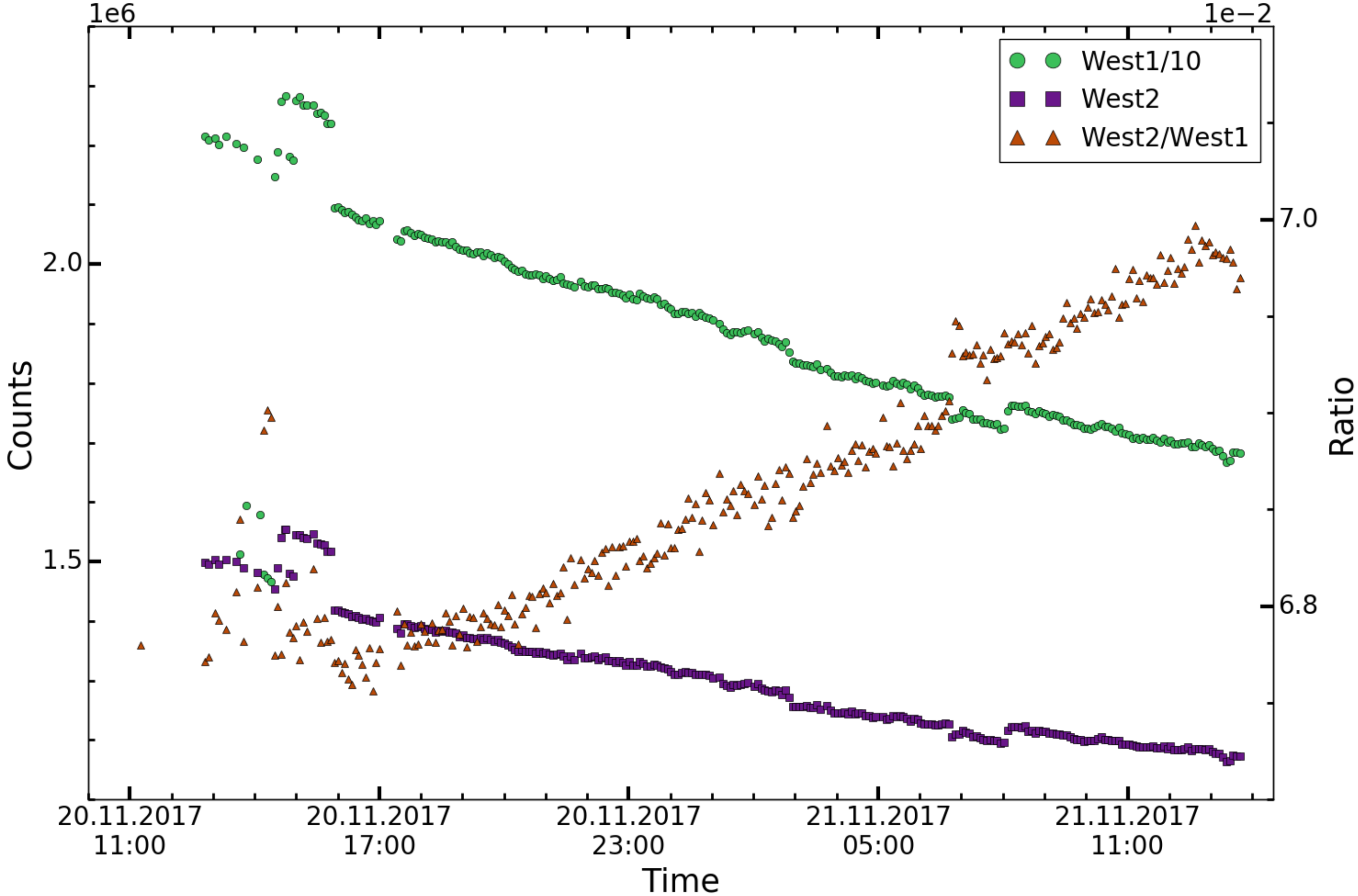
- Similar to $^{1}H_2$, D_2 has two spin isomers called ortho and para D_2
- D_2 is a homonuclear diatomic nuclear and D has integer nuclear spin (ground state S = 1) → system of two undistinguishable bosons → wave function must be symmetric under exchange of the deuterons
- $\Psi_{tot} = \Psi_{vib}\ \Psi_{rot}\ \Psi_{spin}$, where Ψ_{tot} must be symmetric and Ψ_{vib} is always symmetric
- For Ψ_{tot} to be symmetric, the following combinations result

S	Degeneracy	Ψ_{spin}	Ψ_{rot}	J	State
0	1	Symmetric	Symmetric	Even	Ortho
1	3	Antisymmetric	Antisymmetric	Odd	Para
2	5	Symmetric	Symmetric	Even	Ortho

- Ortho states more stable than para, but self-conversion very slow (τ = 80 days)
- In terms of UCN production, a high para content leads to a high number of para to ortho conversions through interaction with UCN, resulting in a high increase in kinetic energy of the neutron and effectively eliminating the UCN

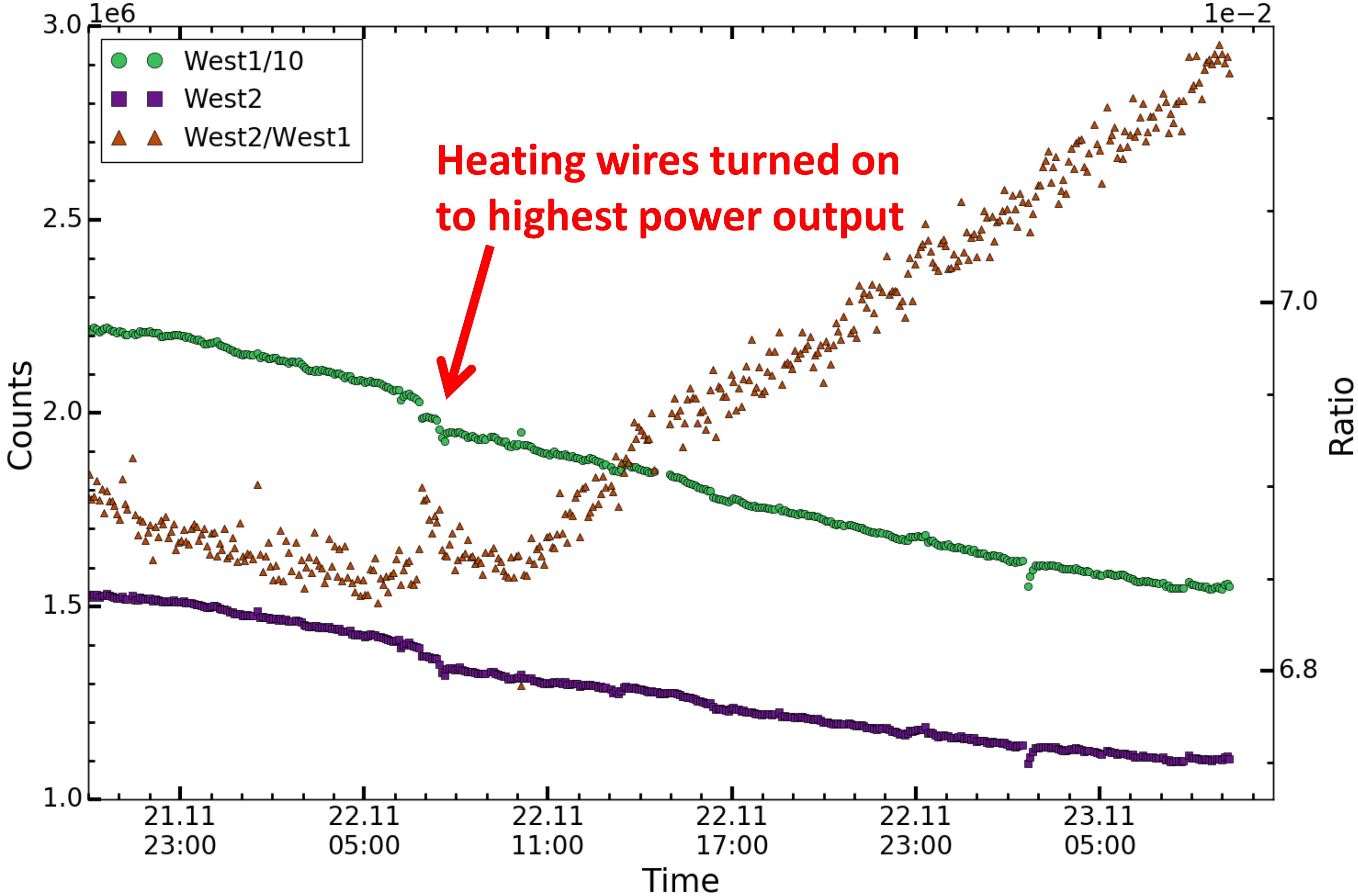
Further Examples of Ratio Change

- Example of West2/West1 ratio change with all port shutters open



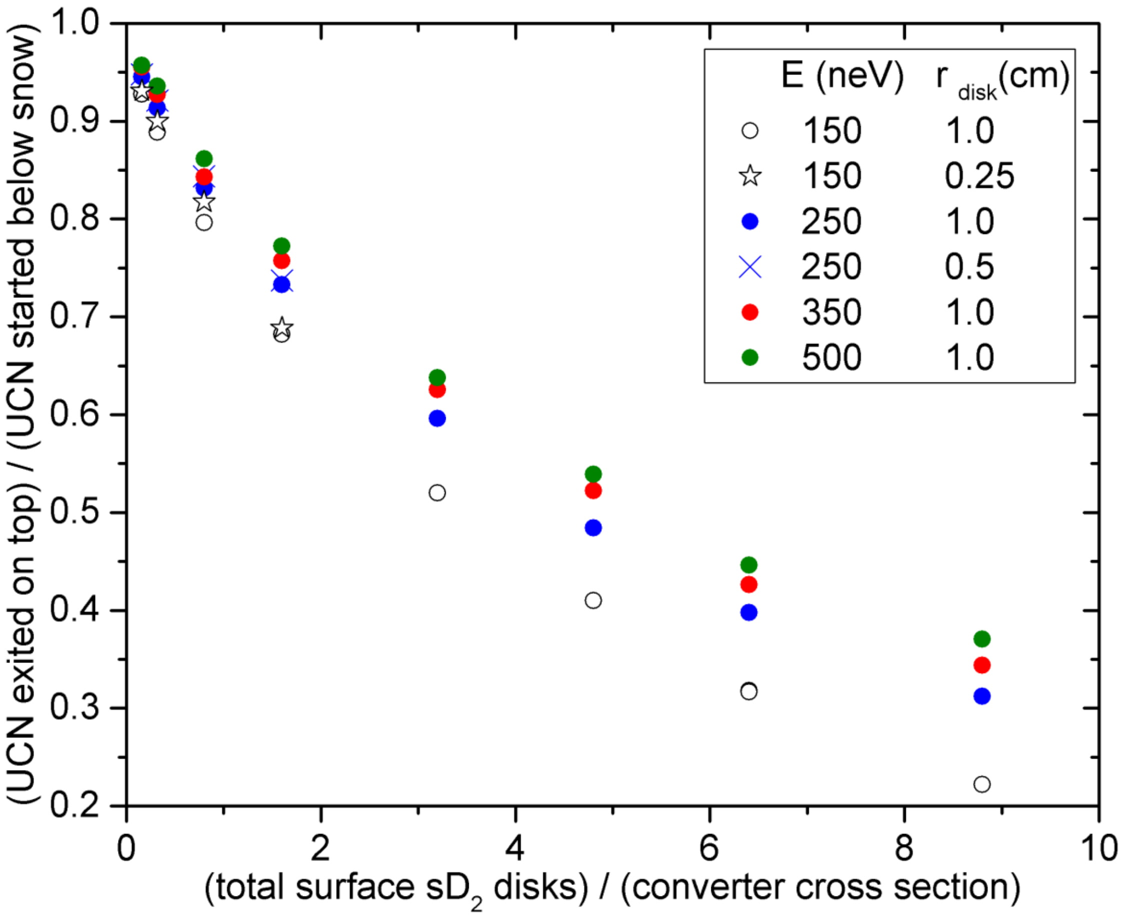
Further Examples of Ratio Change

- Lid heating during operation with high cooling power does not eliminate the frost



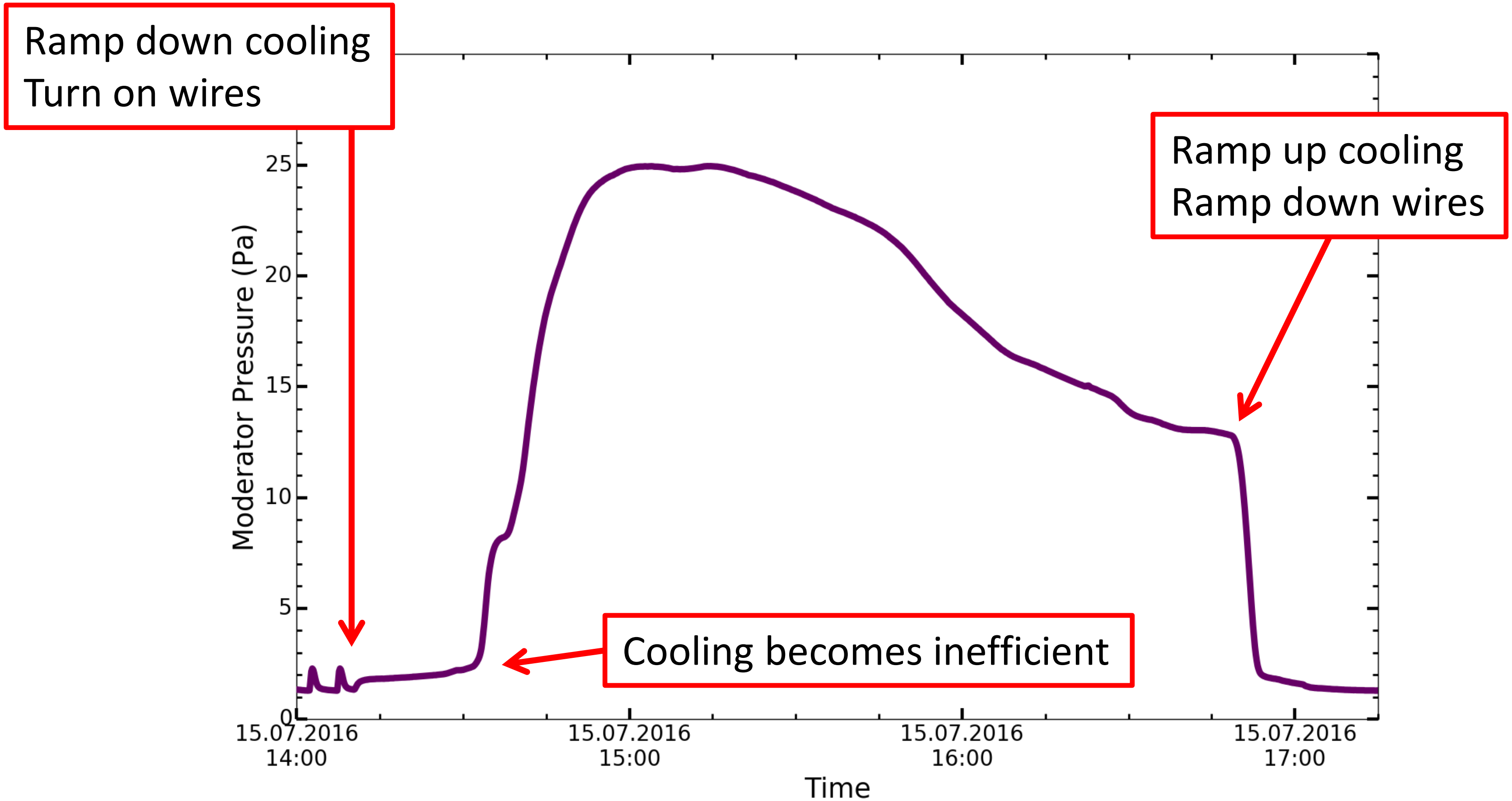
UCN loss due to Frost Disks

arxiv:1804.08616v2



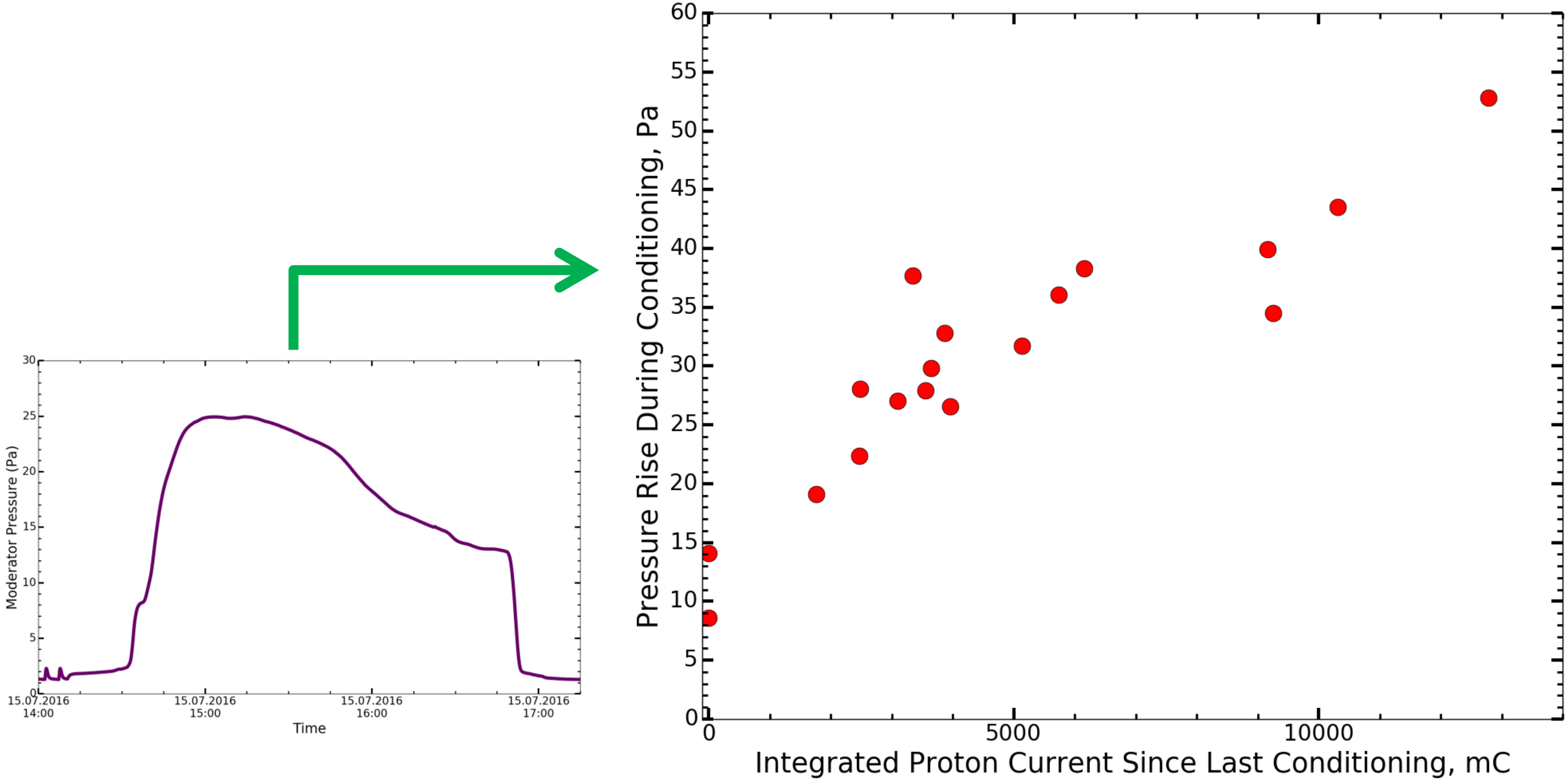
D_2 Pressure during Conditioning

- The pressure inside the moderator vessel shows a typical evolution during conditioning



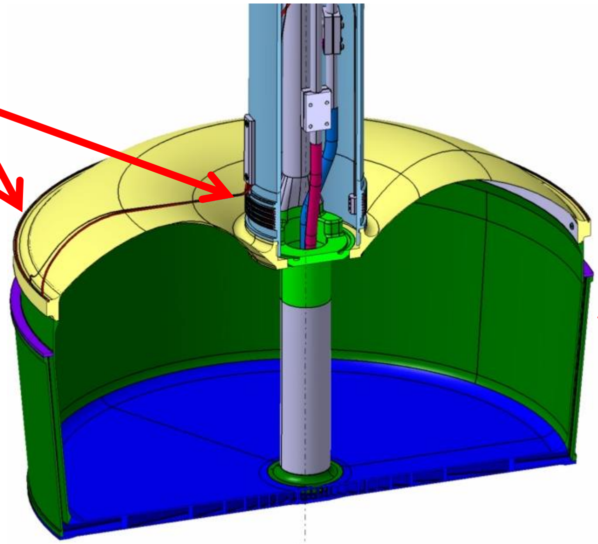
Integrated Current vs Pressure during Conditioning

- **Further evidence for D_2 structures building up during operation**: the more we pulse until the next conditioning, the higher the D_2 pressure during conditioning

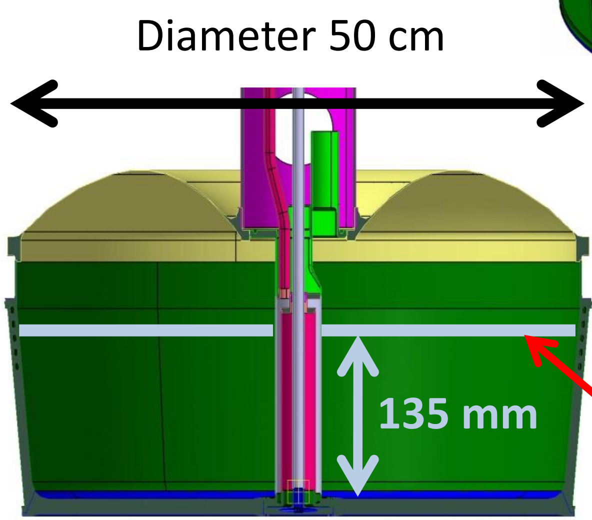


Cold Moderator Vessel

Lid Heating wires, 15 W each



Moderator vessel, $AlMg_3$, total allowed sD_2 volume of 30 L, cooled with supercritical He



Fill height for 4.7 kg, i.e. 23 L sD_2 at 5 K

Coolant He flow