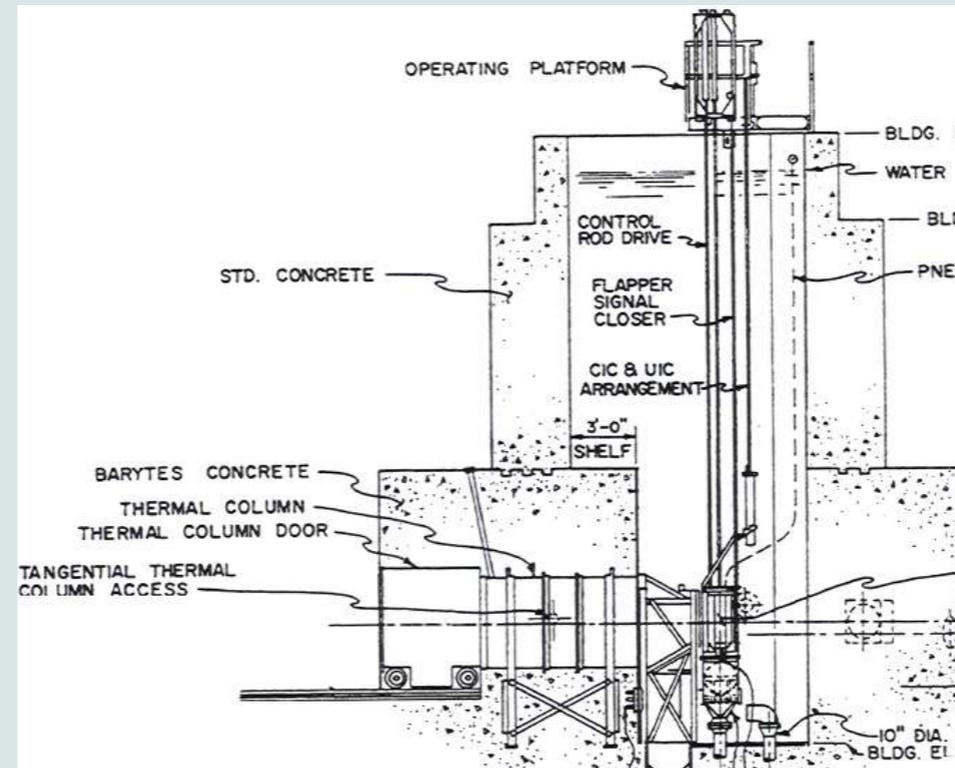


# PULSTAR UCN source: studying solid deuterium growth and evolution



***E. Korobkina, G. Medlin, C. White, E. Lutz  
A. Young, B. Wehring, P. Huffman, A. Hawari, R. Golub***

***NC State University***



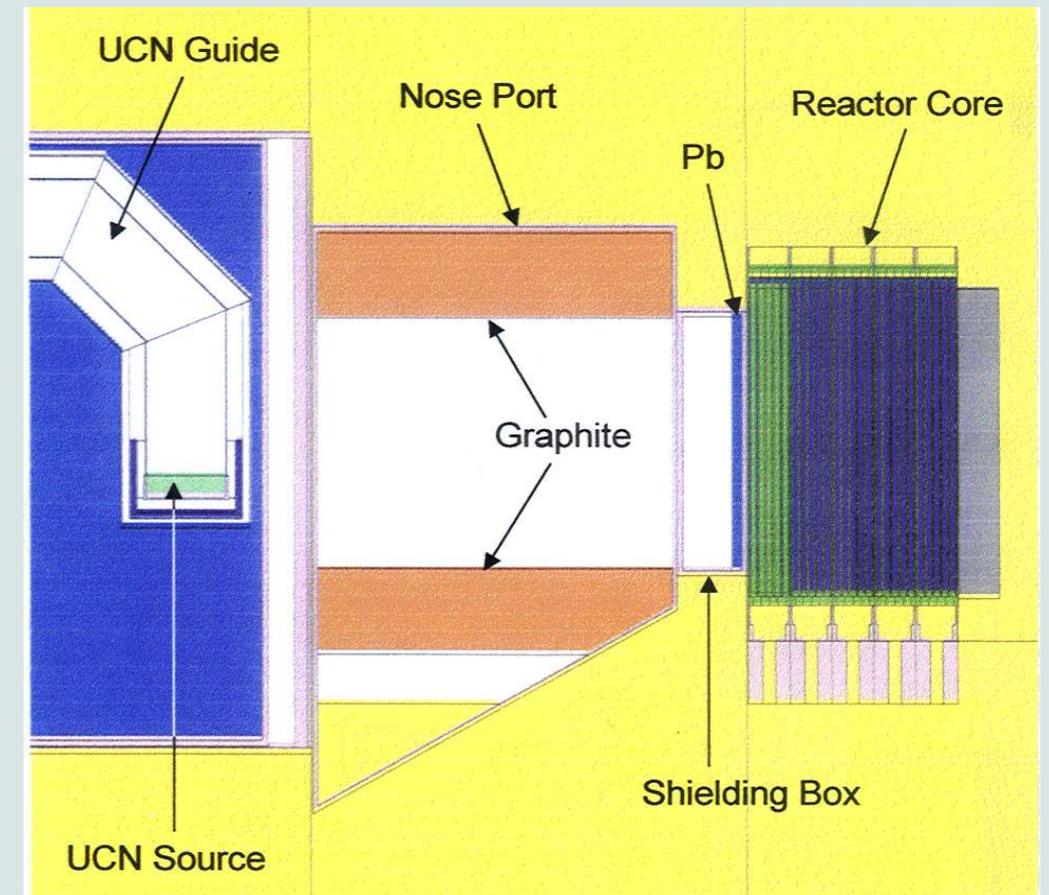
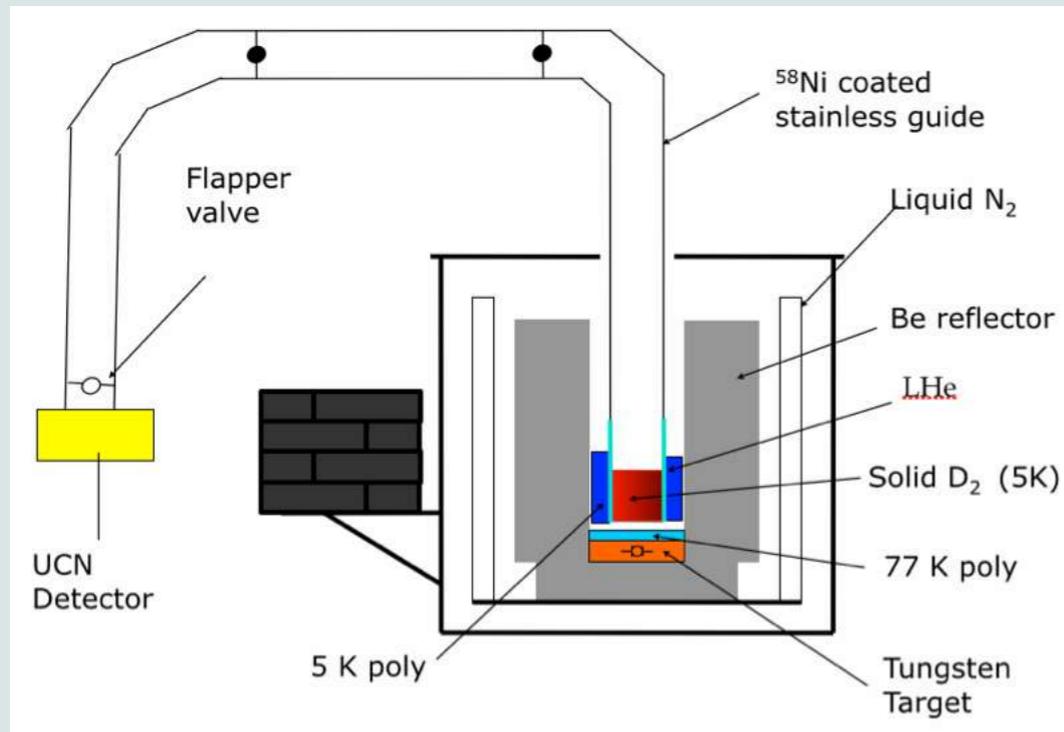
# Conceptual design

LANL pulsed prototype source:

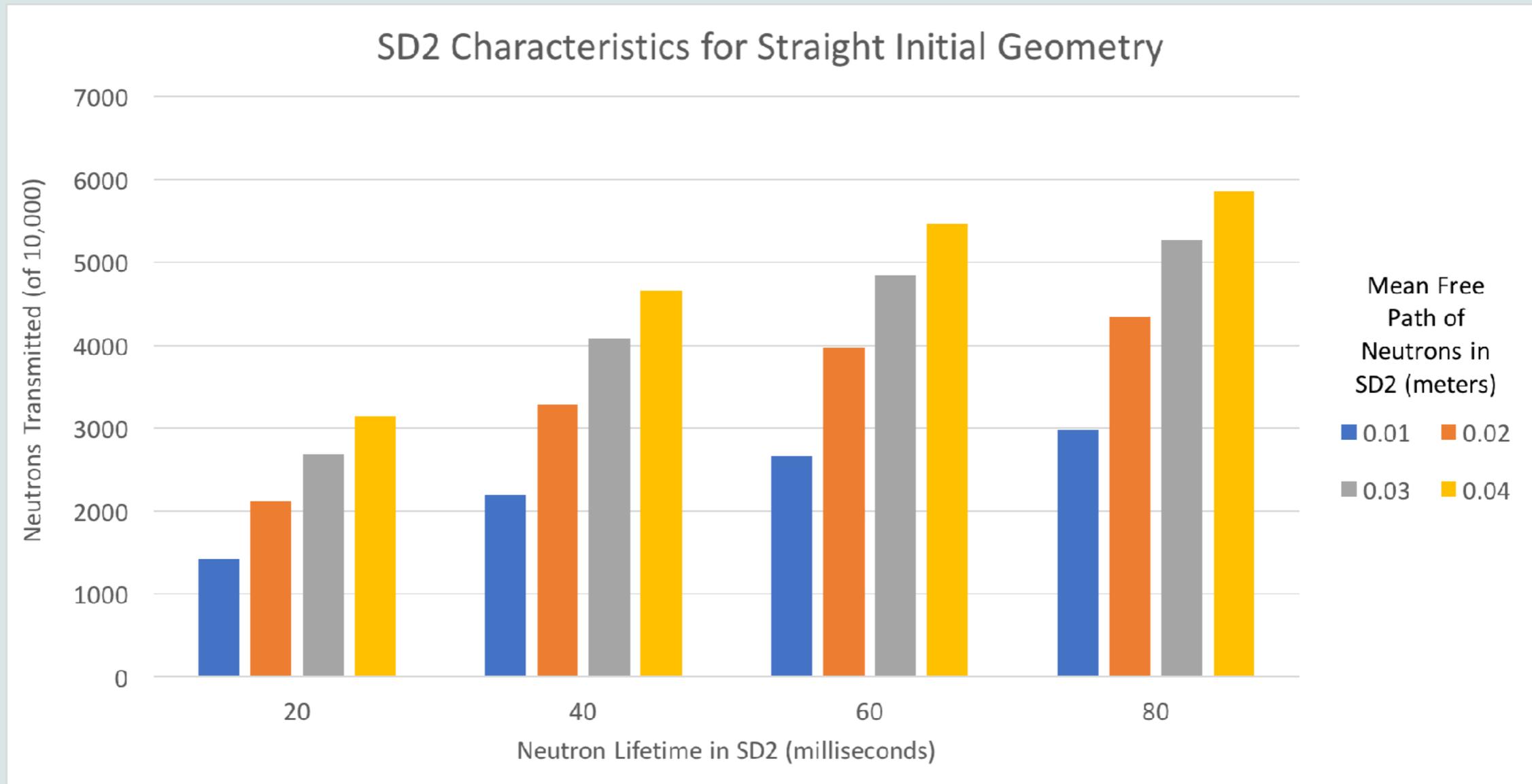
- proton+tungsten as a neutron source
- Be reflector
- 5K & 77K Poly as pre-moderators
- D2 crystal grown from vapor
- 120 UCN/cc in horizontal guide measured
- LHe dewar

PULSTAR conceptual design (2005)

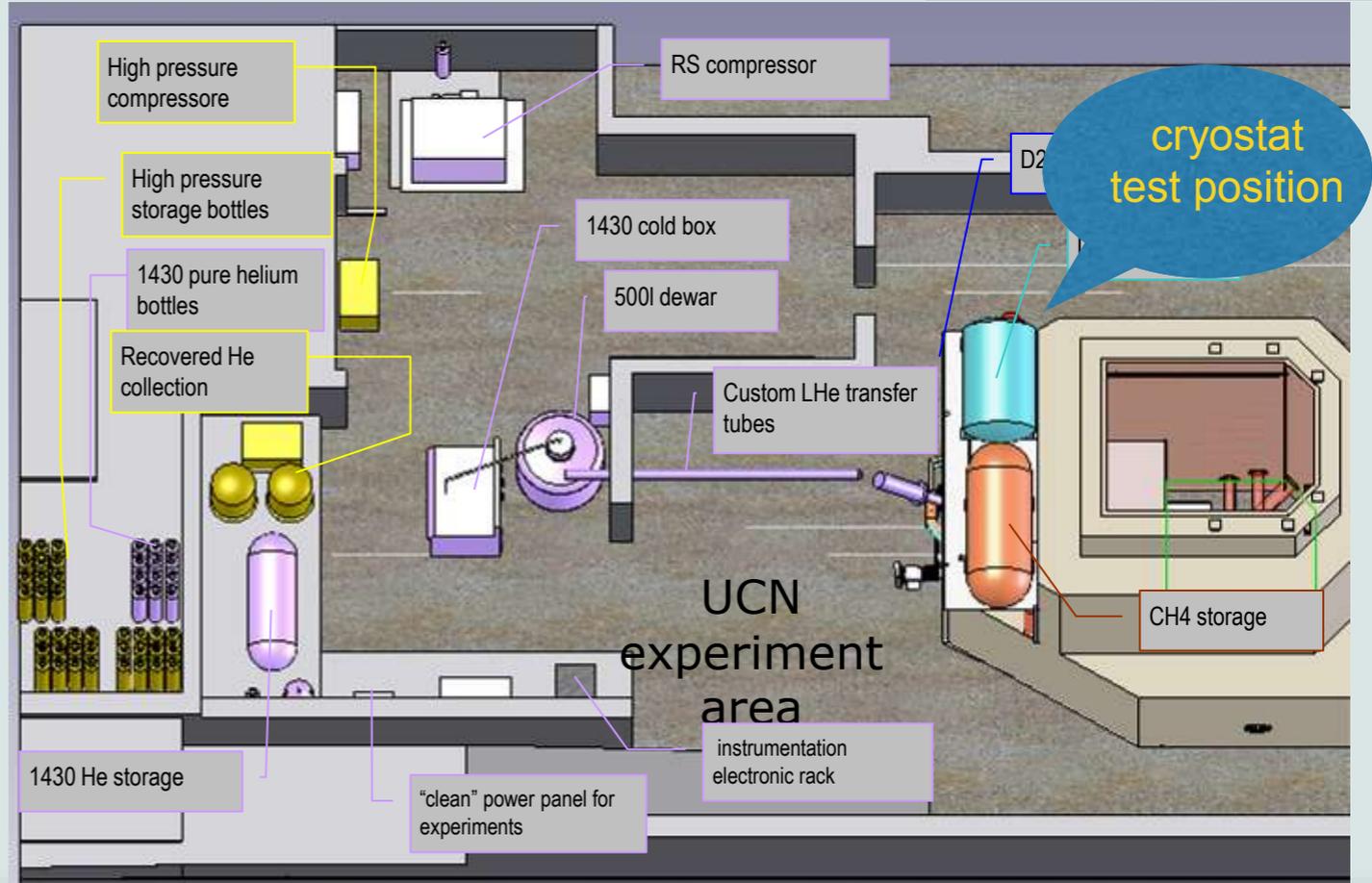
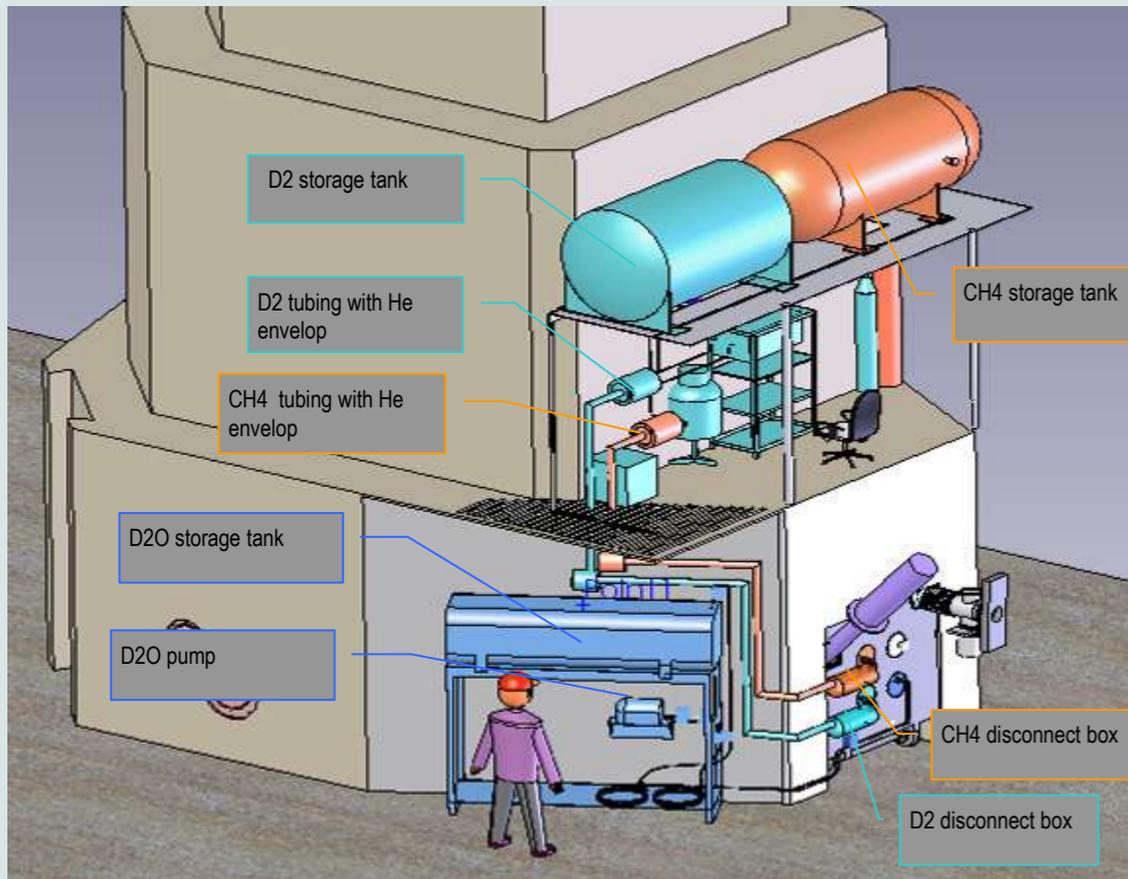
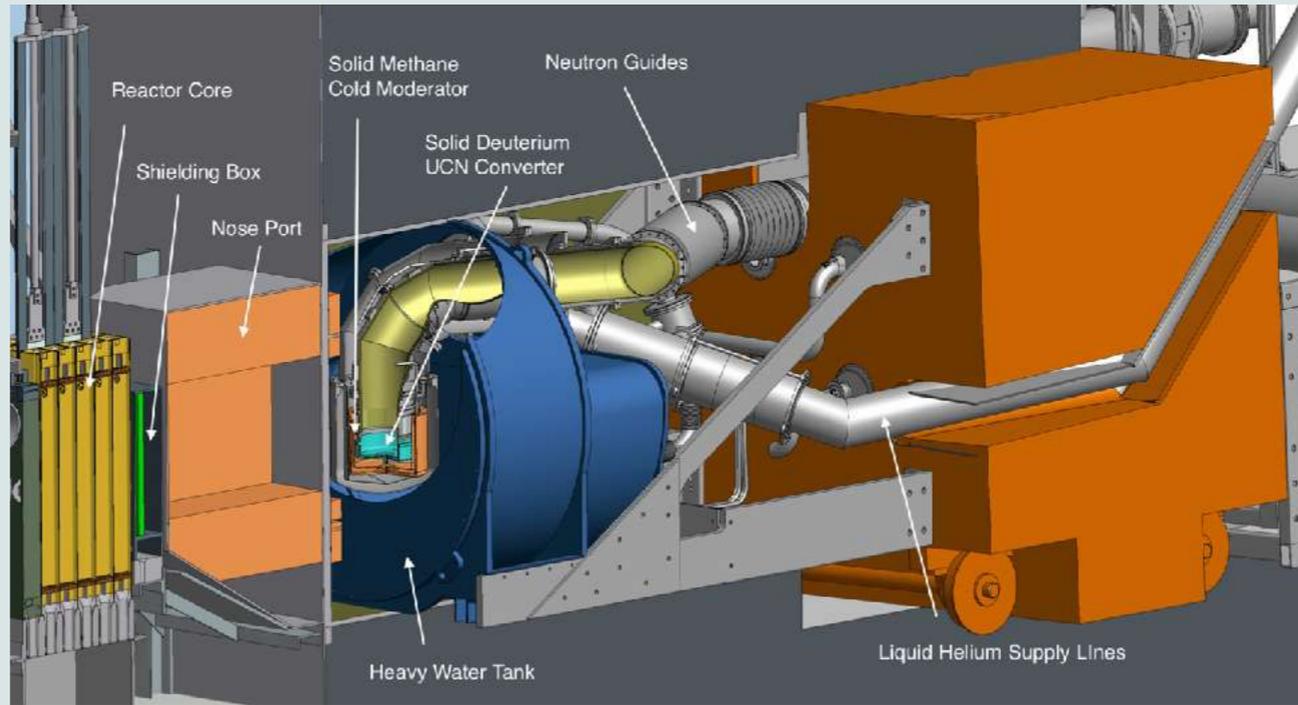
- reactor as a neutron source
- Graphite reflector
- D2O (300K) and solid methane (~40K) as pre-moderators
- D2 crystal can be grown from vapor
- ~ 30 UCN/cc at the exit port predicted
- flow of LHe



# Discussion: MC Simulations of Frost vs Roughness



# Engineering PULSTART source design



# SD2 monitoring system for visual control

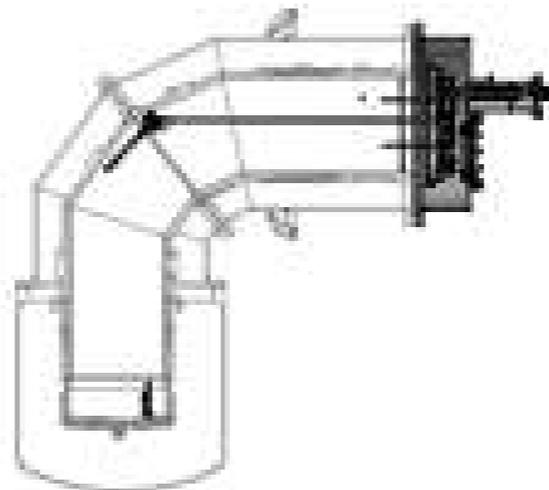
- Graham Medlin designed and implemented a monitoring system which allows to observe D2-container by using camera outside cryostat



Introduction
Neutron trapped mode
Signal transmission
Crystal growth study

## Design

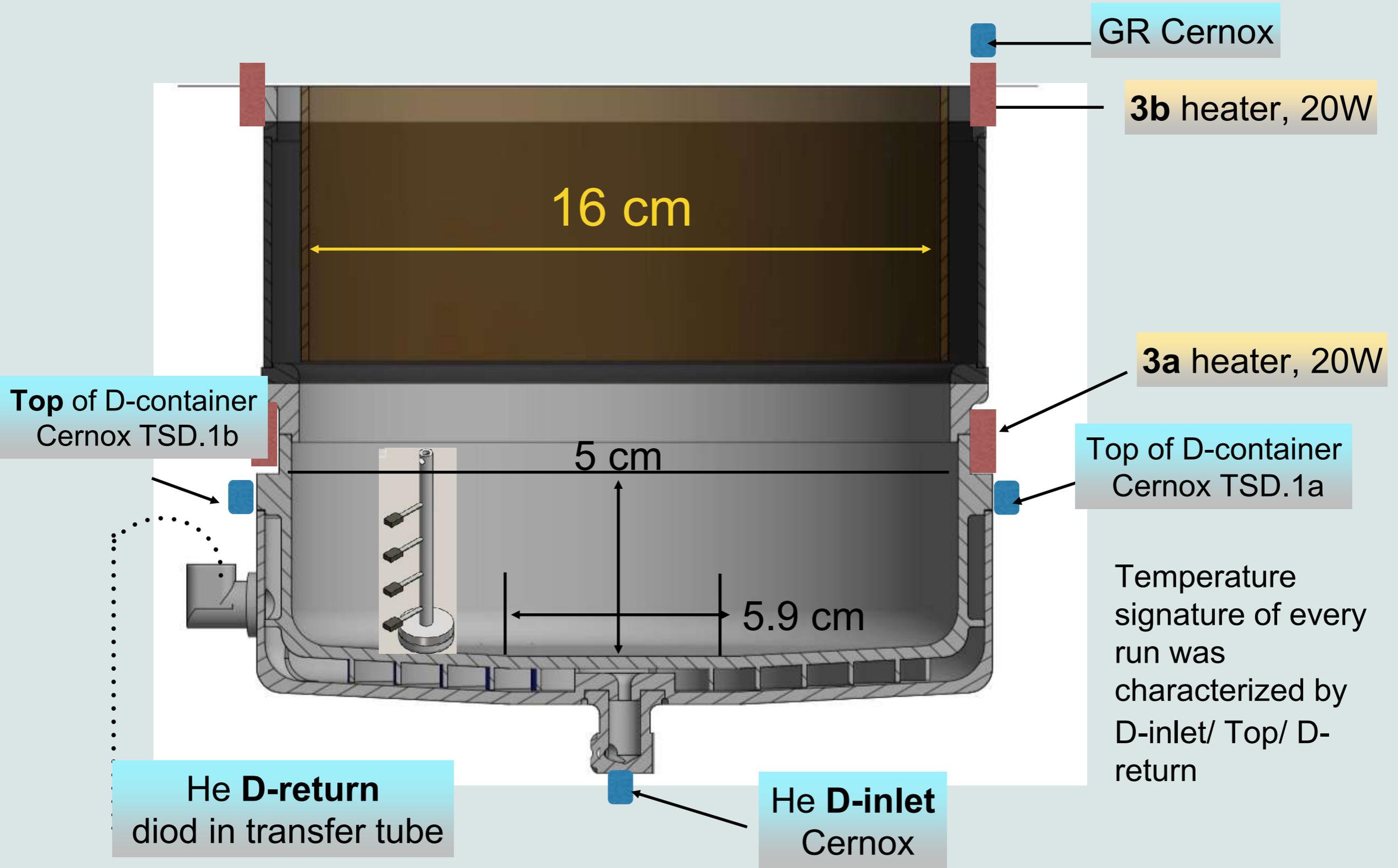
- ▶ Re-usable, install without disassembly
- ▶ Replace UCN foil window
- ▶ Pressure- SS bellows feedthrough
- ▶ Optical- "Dentist's mirror"
- ▶ IR- heated plate
- ▶ T-gradient- stand lowered from mirror





G. Medlin
Pulsed Ultrasound Neutron Source
NC State

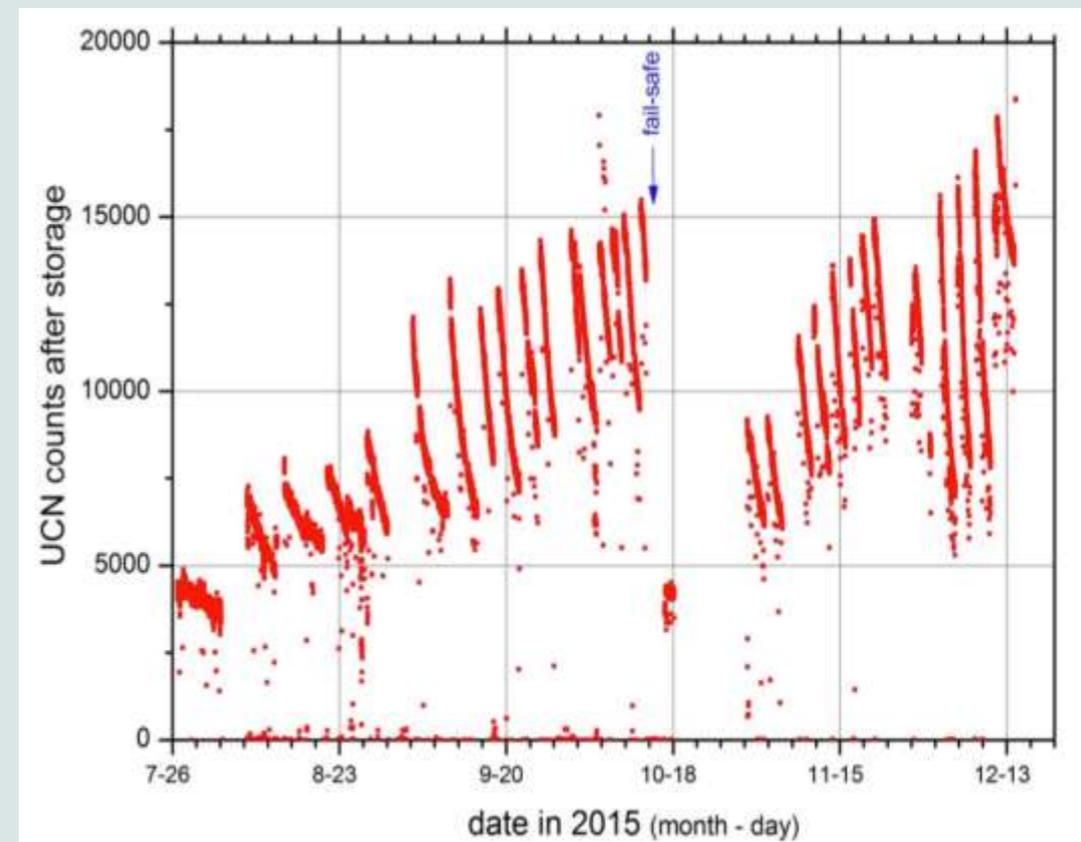
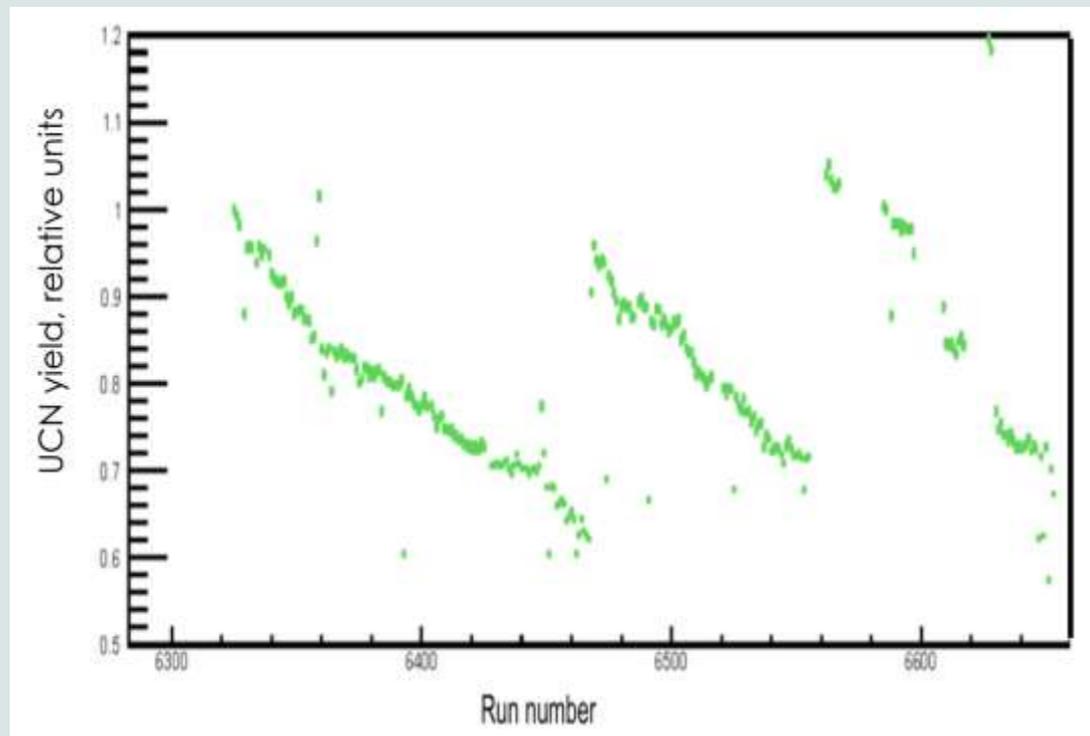
# SD2 growing tests : instrumentation details



# Goals of our study



1. Learn how to growth of SD2 in real UCN source cryostat, as it will be done when source will be in operation
2. Study effect of Temperature pulsing on the originally transparent SD2 crystal to understand UCN yield degradation on the pulsed UCN sources (LANL, PSI, Mainz without moderator)



# Overview of runs

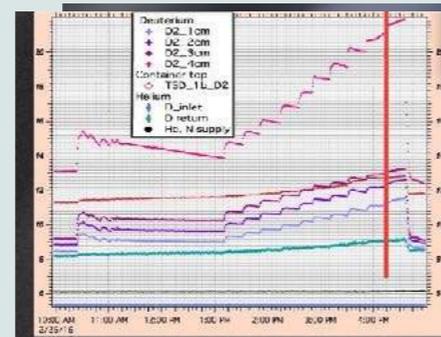


1. Feb 23-26 **condensation from vapor**
2. Feb 27-Mar 4 - annealing, **melting-refreezing**; evaporation
3. Mar 15- Mar 25 - **condensation from vapor**
4. Mar 25 - 28 annealing, adjustment of cryogenic system
5. Mar 29-31 **heat shooting**
6. Apr 1-4 slow warming/annealing ; evaporation
7. April 11-13 **condensation from vapor**
8. April 25 **condensation from vapor**

# Feb 23-26 condensation, 5.4K/ 8.2K/ 7.2K:



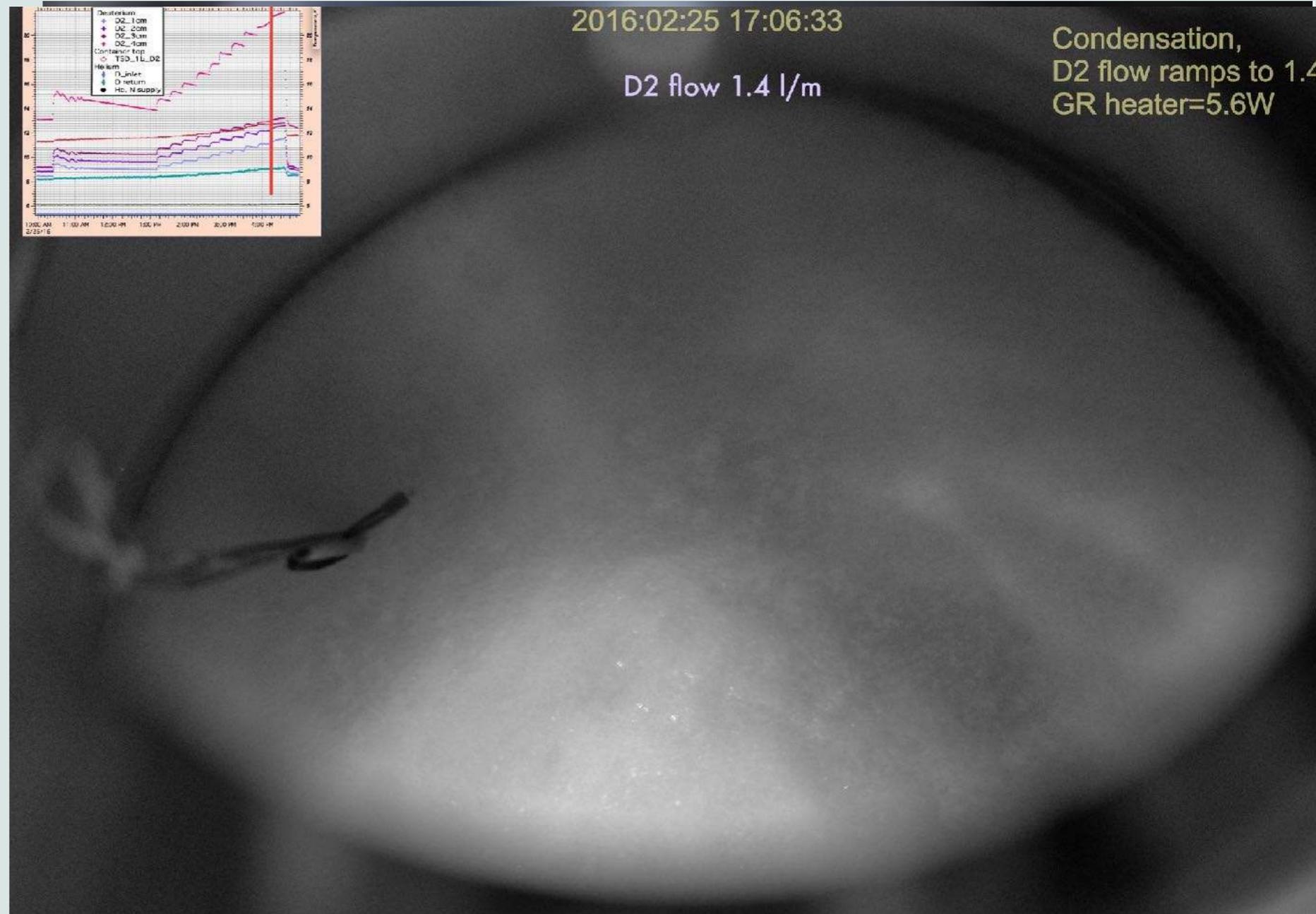
- this run was to simulated Mainz UCN source condensation of SD2 with cold (6K) bottom of container and slow D2 flow rate
- Small flow (0.3 l/m) produced dense multicrystall, optically opaque
- Higher D2 flow >1 l/m produced snow-flake-like mass
- when T of container was increased (**5.4K/ 11.2K/ 8.3K** ) and crystal annealed, D2 flow =>0.8 l/m results in visibly shiny surface



2016:02:25 17:06:33

D2 flow 1.4 l/m

Condensation,  
D2 flow ramps to 1.4  
GR heater=5.6W



# Feb 27-Mar 4 - melting-refreezing

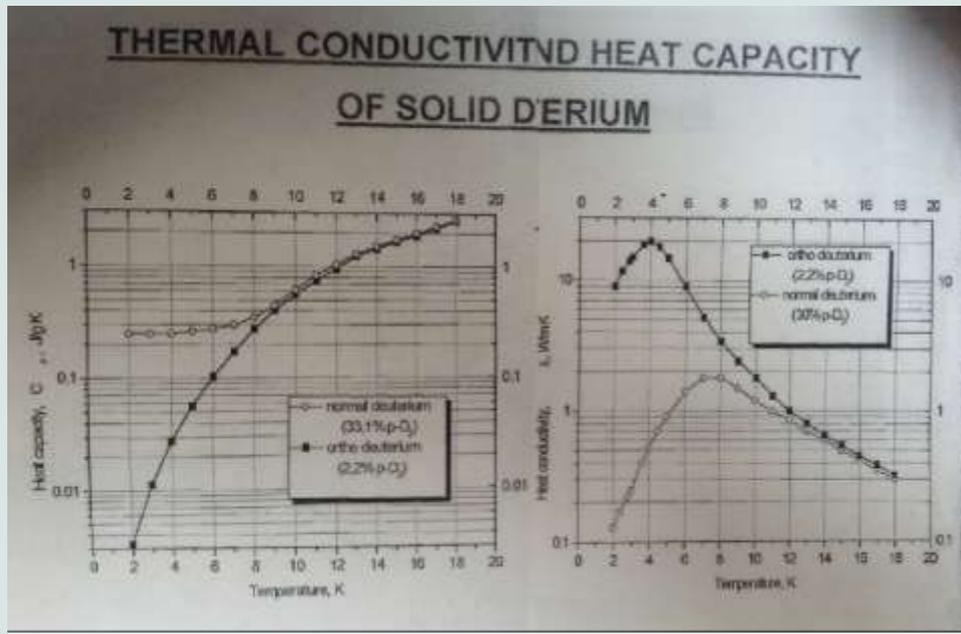
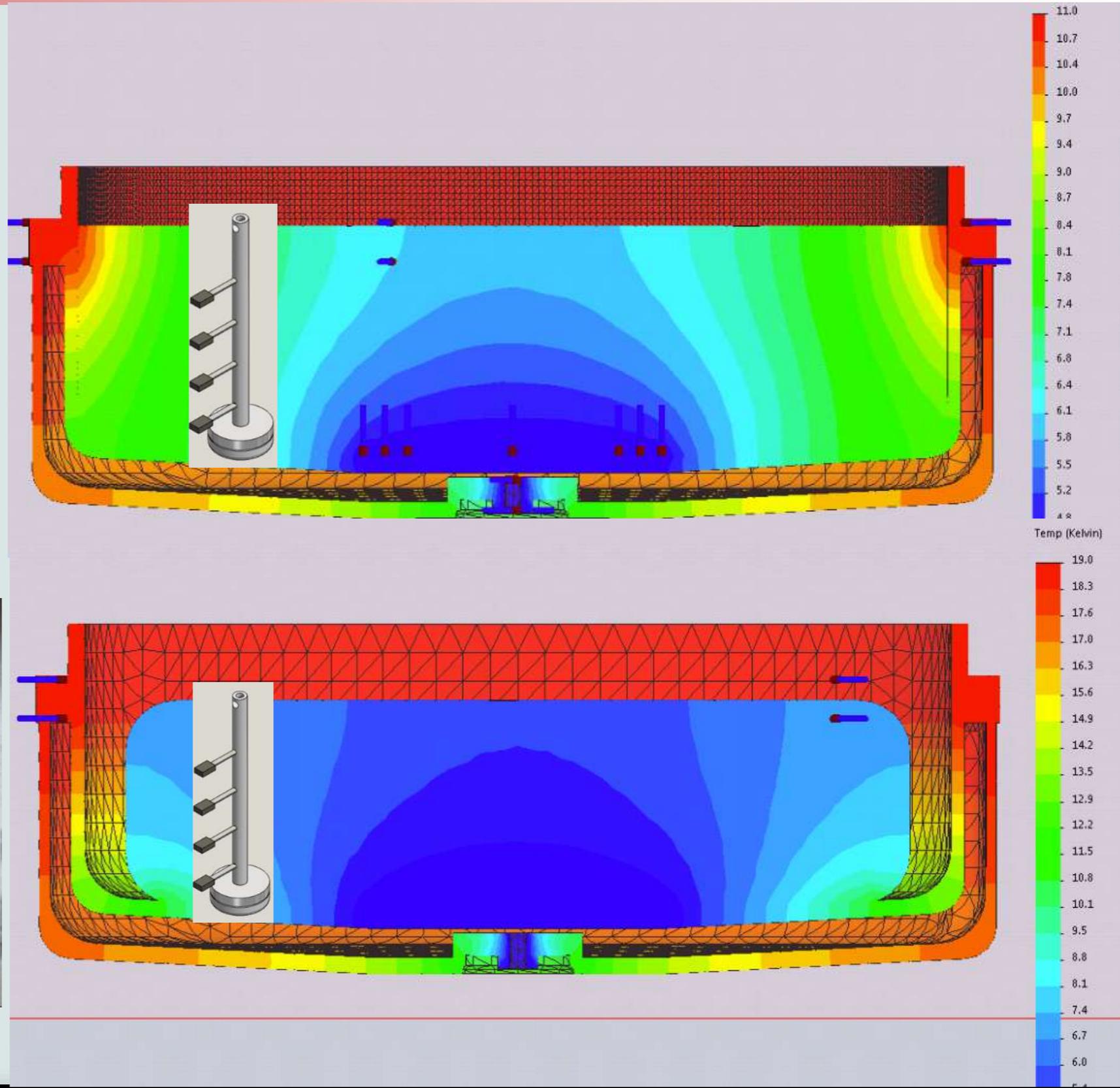


- Melting & re-freezing is routine crystal conditioning procedure at LANL and PSI UCN source
- it produces reasonable transparent crystal, with some crystal structure visible, no surprises here
- After cooling down to 5K (inverse T distribution), this crystal was melted and evaporated



# SD2 growing test: complete cool down

From liquid	
liquid He cooled (K)	
3/3 14:00-16:00	
8.244	top of container
6.229	He-inlet
5.103	4 cm
5.190	3 cm
5.137	2 cm
5.368	1 cm



# SD2 growing test 1, conclusions

- with cold bottom around 6K and flow of 0.4 l/m a dense and milky multi crystal is growing
- under the same conditions, except the flow was increased to 1-2 l/m, D2 sublimates as fluffy snowflakes
- melting and re-freezing results in visually quite good crystal

**r container to grow transparent crystal from gas phase - sta**

# Mar 15- Mar 25 - condensation/conditioning



- Mar 15 condensation, no heaters, small cooling power, **9/ 17.5/ 17.5, D2 flow 0.8 l/m:**

- ideal transparent crystal

- Mar 15-16-18, evolution to blob at 11.5K:

- amazing mobility and tendency for avoiding warmer surfaces

- Mar 16 condensation, with heaters on, 8.5/ 18/ 15K, 1 l/m



- **Conclusions:**

- optically clear crystal
- high mobility and tendency to pull off the foreign objects

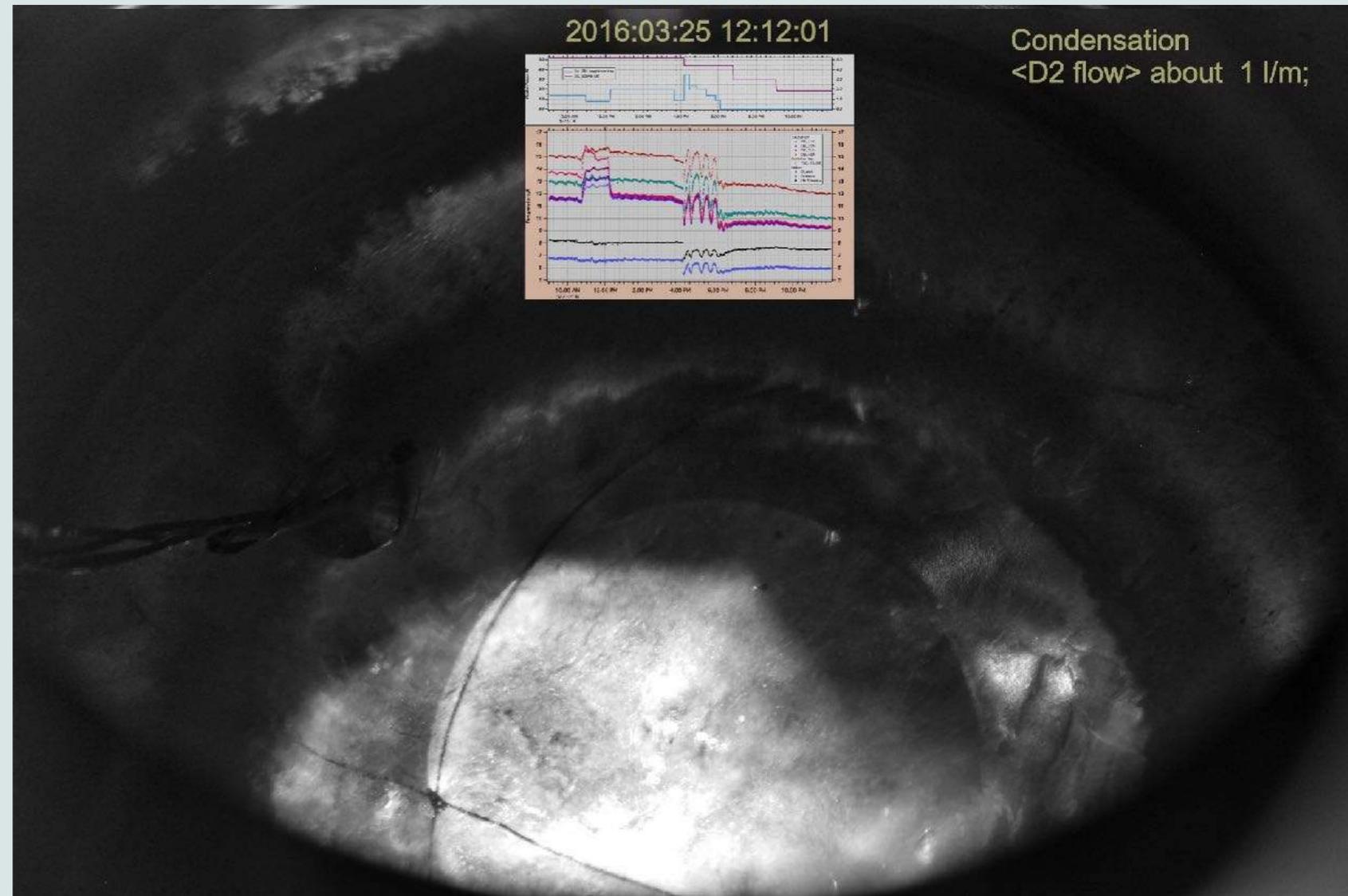
# Mar 22- Mar 25 - condensation/conditioning



- Mar 22 -25  
condensation,  
with heaters, **7.75/ 17/  
18.5-19, flow 1 l/m**
  - transparent crystal
  - all probe sensors  
covered, about 1050  
cc total

- **Conclusions:**

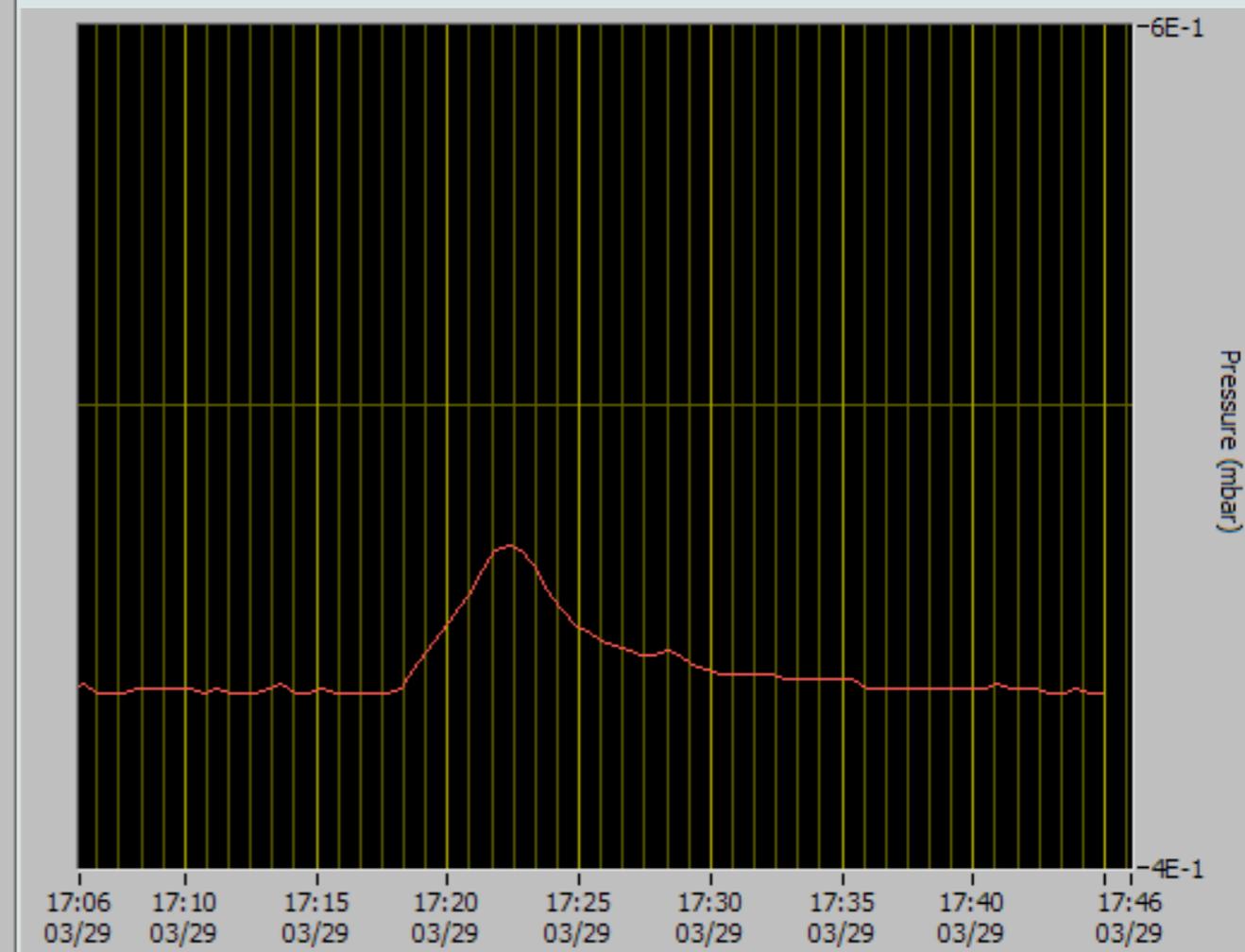
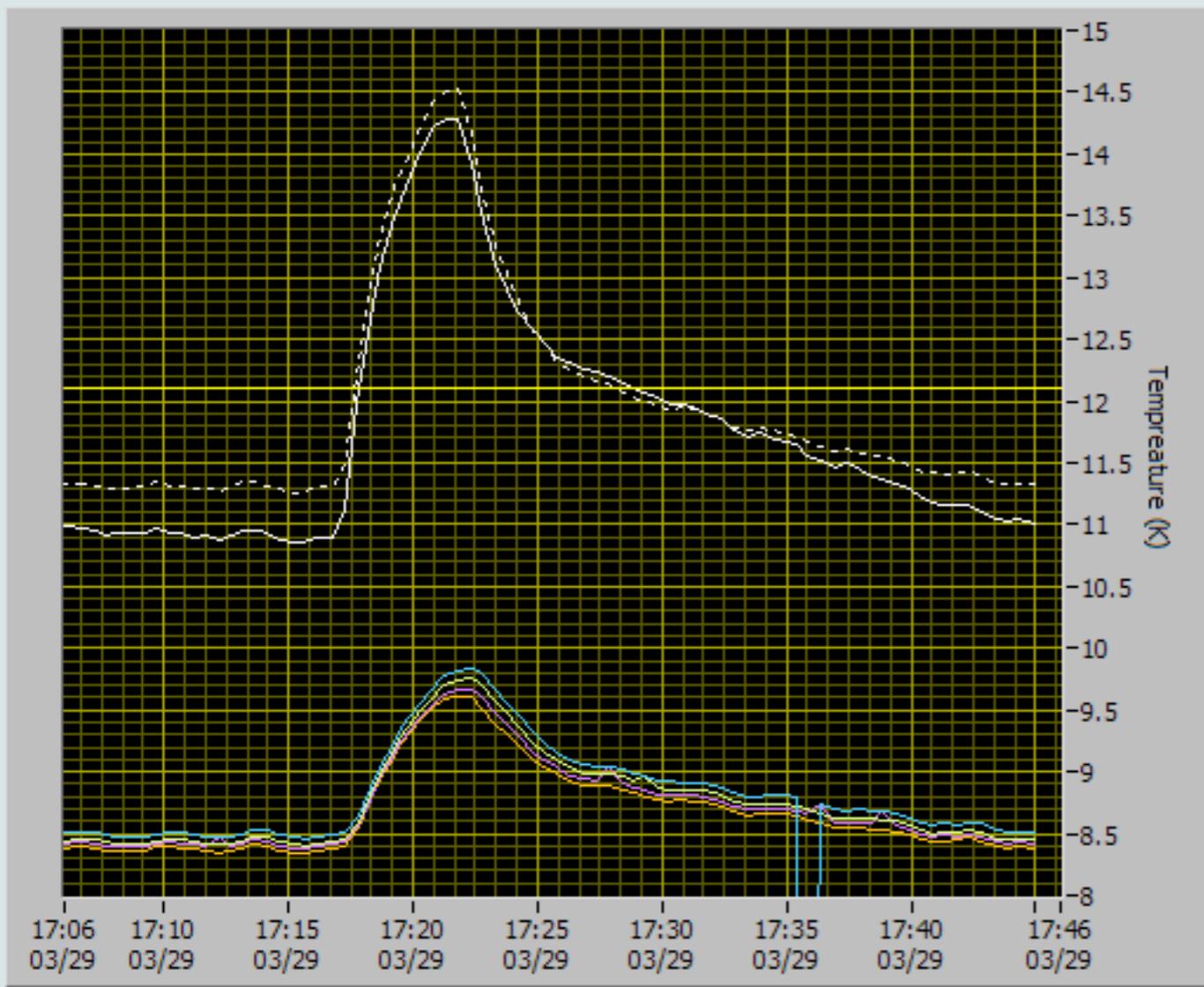
- it is possible to grow  
optically good crystal  
from vapor at warmer  
temperatures and flow  
0.8-1 l/m
- need to be concerned  
about crystal shape  
evolution with time



# Mar 29-31 heat shooting



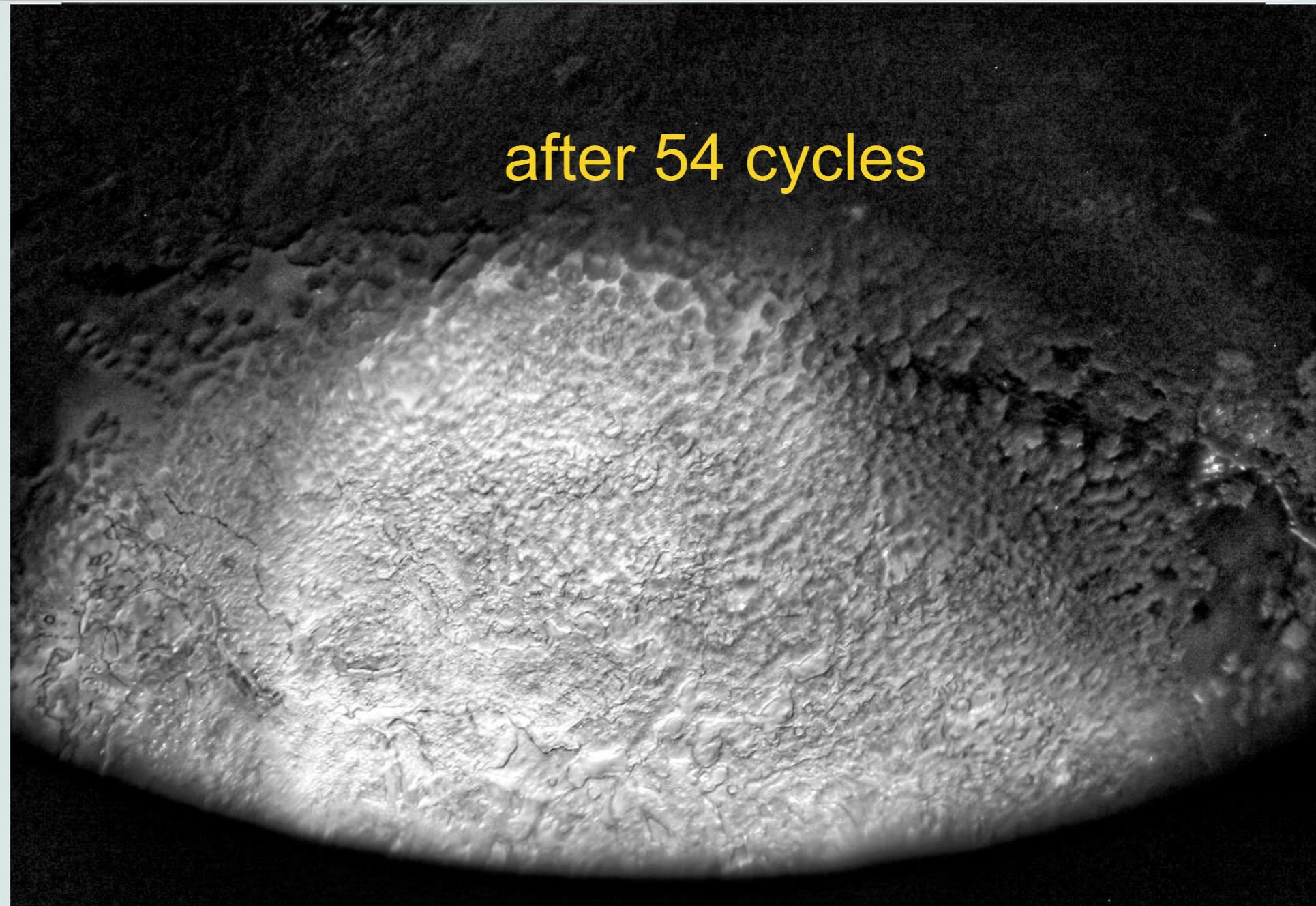
- We used heater to simulate heat input from the proton beam
- Power was chosen to simulate pressure and approximate T-rise from PSI data



# Mar 29-31 heat shooting



- Accidental He flow oscillations on Mar 25
- Mar 29-31 heat shooting
- **Conclusions:**  
we need also to be concerned about crystal surface evolutions when temperature is not stable



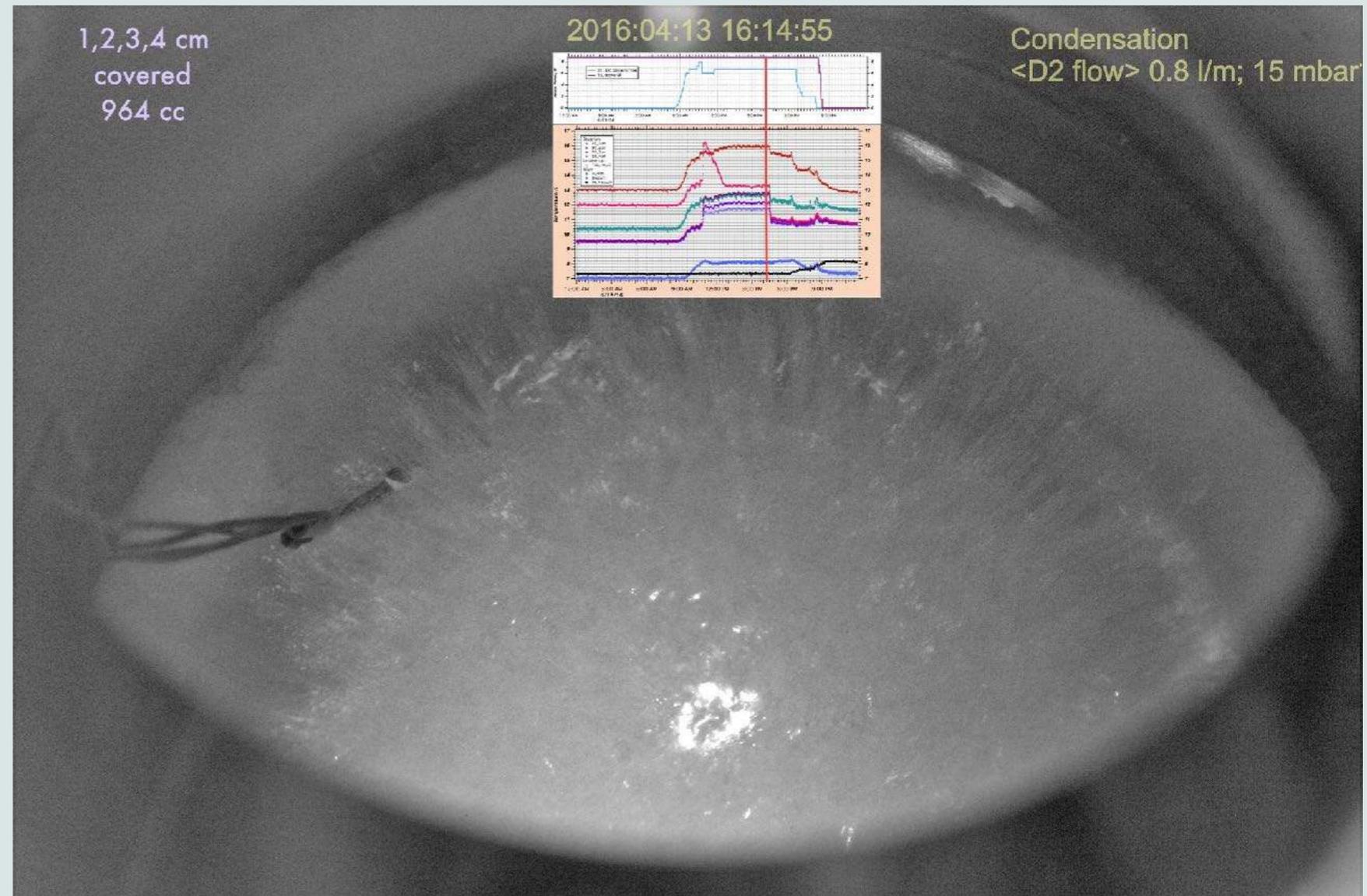
# April 11-13 condensation, 8.3/ 16.2/ 13, 0.8 l/m



- April 11-13 condensation, **8.3K/ 16.2K/ 13K, D2 flow 0.8 l/m**

- **Conclusion:**

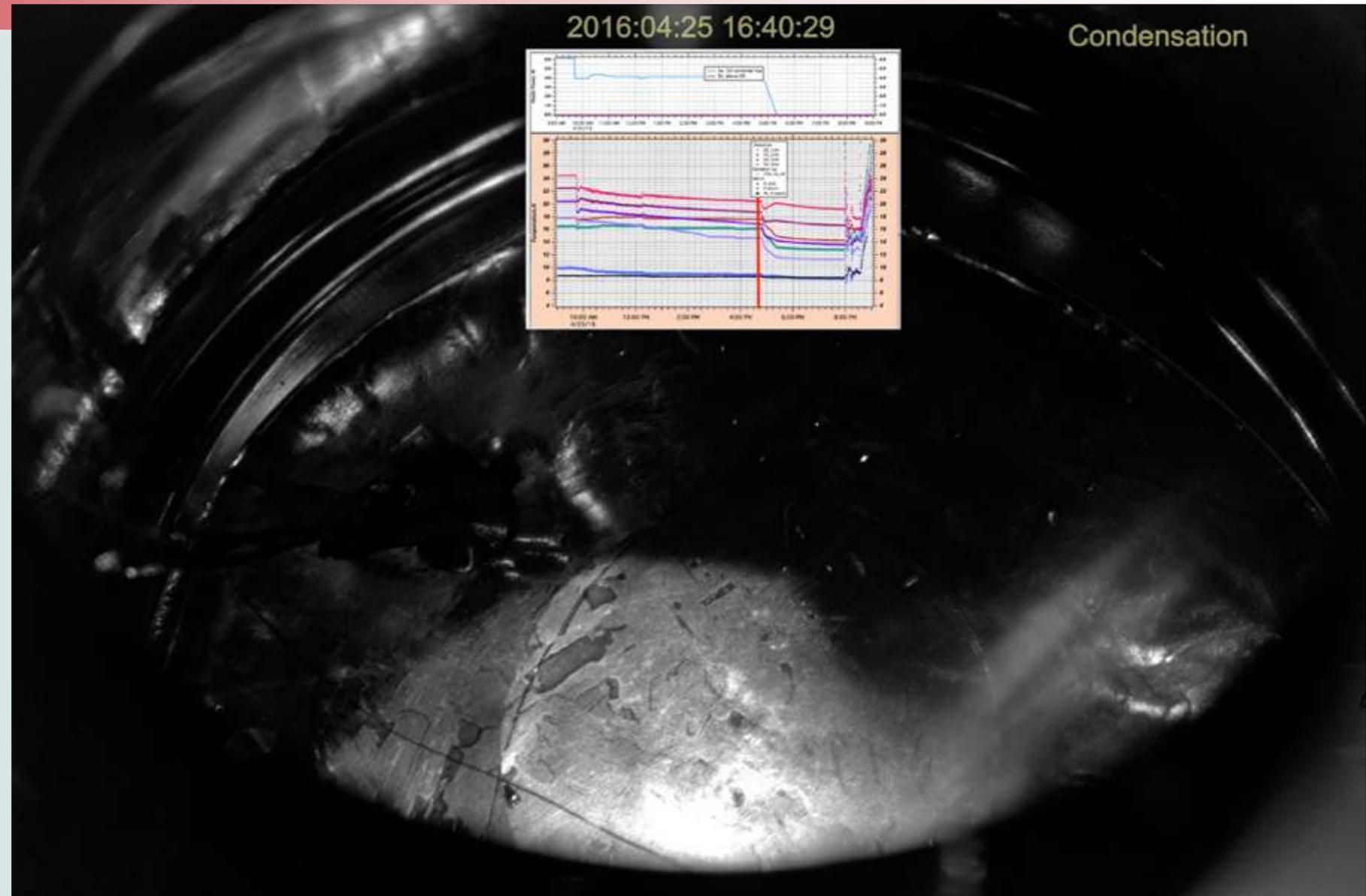
- this attempt to reproduce Mar 15 condensation from scratch, unsuccessful, because D-return was not taken into account and container was too cold
- very interesting run for discussion of crystal properties vs T and crystal shapes



# Lessons learned: April 25 condensation



- April 25 condensation, 10-9/18/ 16.2 , 0.8 l/m
- attempt to reproduce Mar 15 condensation from scratch, quite successful.



- **Conclusions:**

- optically clear crystal
- we need at least  $T > 10K$  at the coldest spot to grow transparent crystal

# Discussion: Growth at low T and avoiding foreign objects



- at  $T < 6\text{K}$  SD2 does not move
- but the growth is very inhomogeneous



# Published studies of SD2 growth

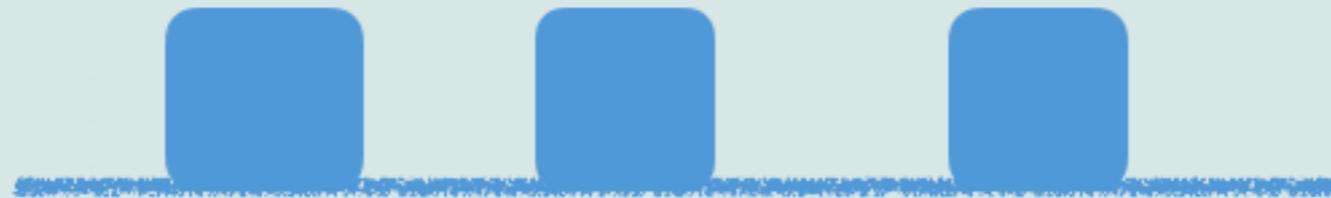
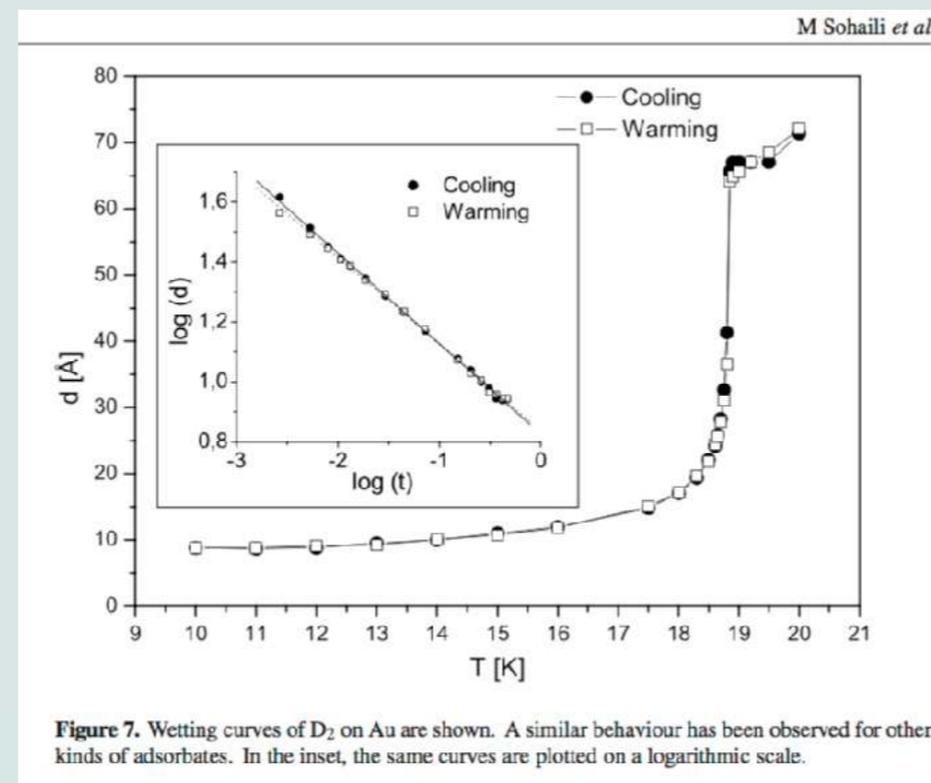


- Solid state physics
  - Study of quantum solids (Physics of quantum solids, Editors V.G. Manzhelii and Y. A. Freiman, AIP, 1997)
- Applied physics
  - SD2 as target for fusion (Hydrogen Properties for Fusion Energy , book by P. C. Souers) - small crystals grown near triple point
  - Search for Neutrino mass in beta decay: Structural Stability of Solid Deuterium Films , 1999, JLTP, vol. 119, 2000, p.615
  - investigation of solid D<sub>2</sub> with respect to ultra-cold neutron sources, study made by PSI group, Dissertation “UCN convertors” by Malgorzata Kasprzak
  - Observation of crystal growth in situ at LANL, unpublished

# Discussion: Growth at low $T < 6K$ and Fast movement from the container surface



- SD2 exhibits so called “triple point wetting”, i.e. it wets surface completely above the triple point, while below only films of several monolayers can be grown and the rest grows as crystallite islands



# Discussion: Fast movement from the container surface, PRL 88 (2002)



- This effect was usually ascribed to the substrate-induced strain in the solid film, which occurs due to the lattice mismatch and the strong van der Waals pressure in the first few monolayers. In 2002 PRL 88 publication, A. Esztermann et al. proposed another reason for “triple point wetting ” : surface roughness

In conclusion, we have shown by theory and experiment that incomplete wetting of solid films on substrates is dictated by the surface roughness rather than by the solid strain caused by the substrate attraction. A finite roughness always enforces triple-point wetting. The theory is in good quantitative agreement with our experimental data. As any real substrate is rough, the described scenario plays a decisive role in any wetting situation where solid layers are involved such as for coatings of sculpted substrates or curved nanoparticles [21,22]. Our results imply that extremely smooth substrates are necessary to generate thicknesses of adsorbed van der Waals films larger than 10 nm which is of direct relevance in such different areas as laser fusion [23], optical spectroscopy [24], surface investigations by slow muons [25], and the determination of the neutrino rest mass [26].

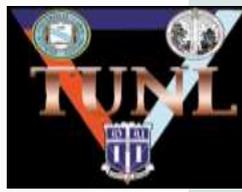
# Discussion: Fast movement from the container surface, PRL 88 (2002)



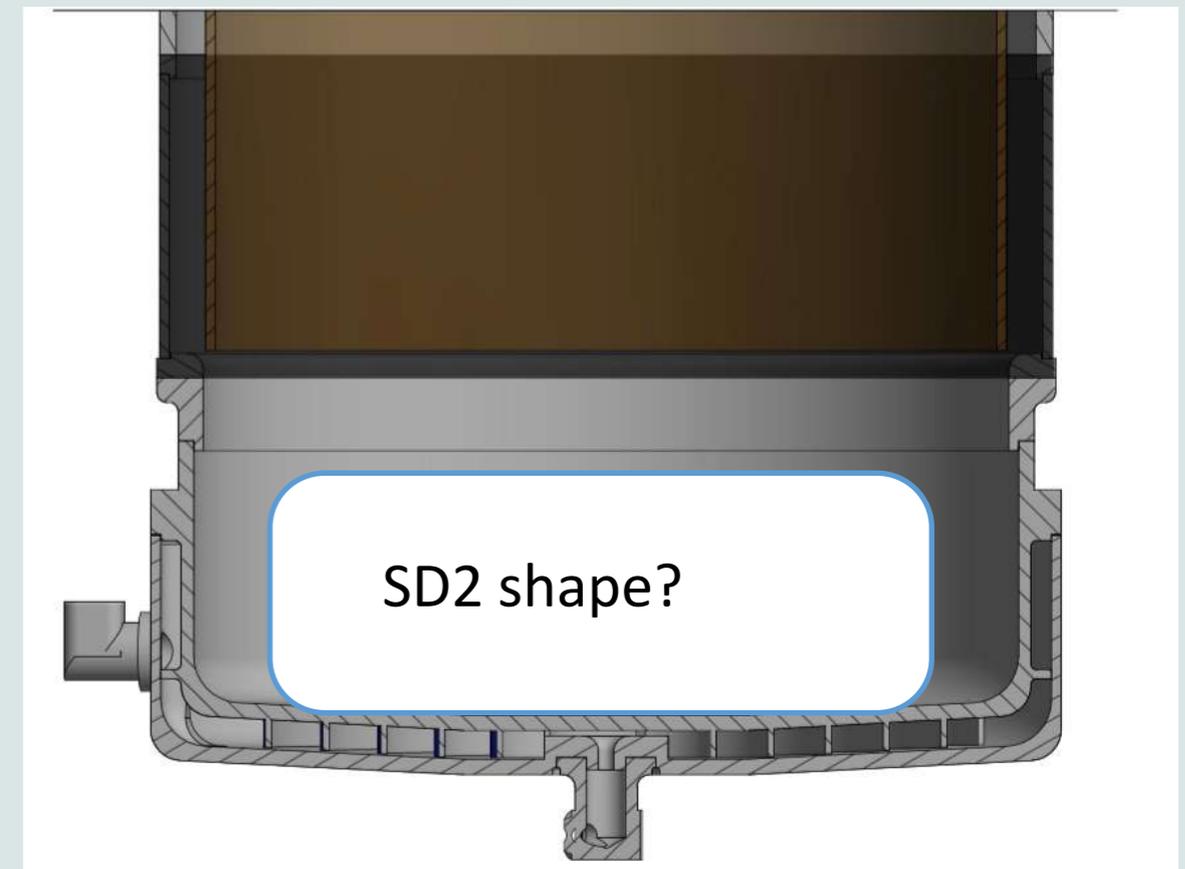
- in contrary to the heat pulsing, there is no frost formation when SD2 is moving around at constant T
- from NMR study: above  $>10\text{K}$  the vacancy induced molecular motion is the main relaxation factor; below  $10\text{K}$  diffusion can be ignored



# Discussion: Temperature dependence of Crystal shapes and transparency



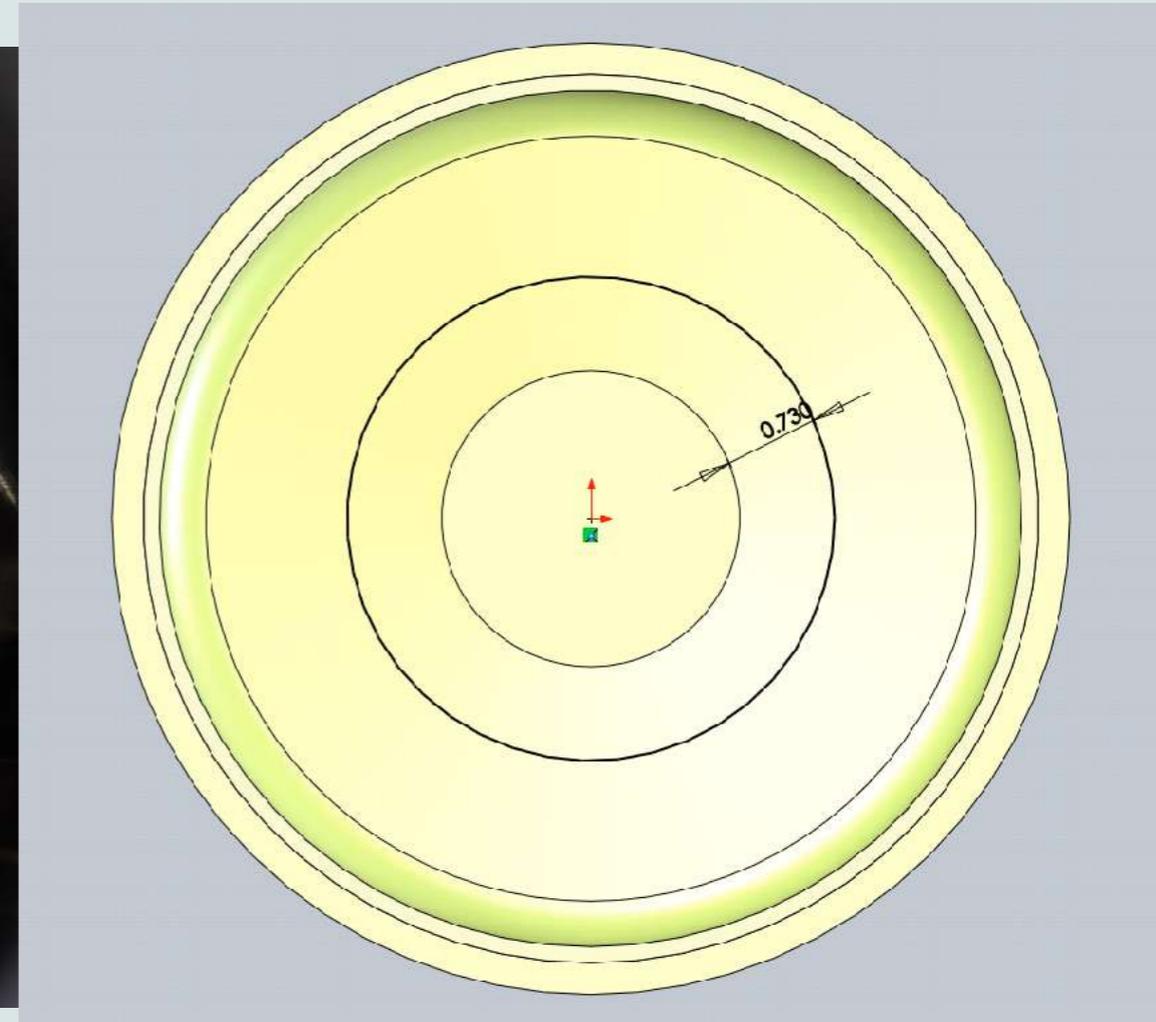
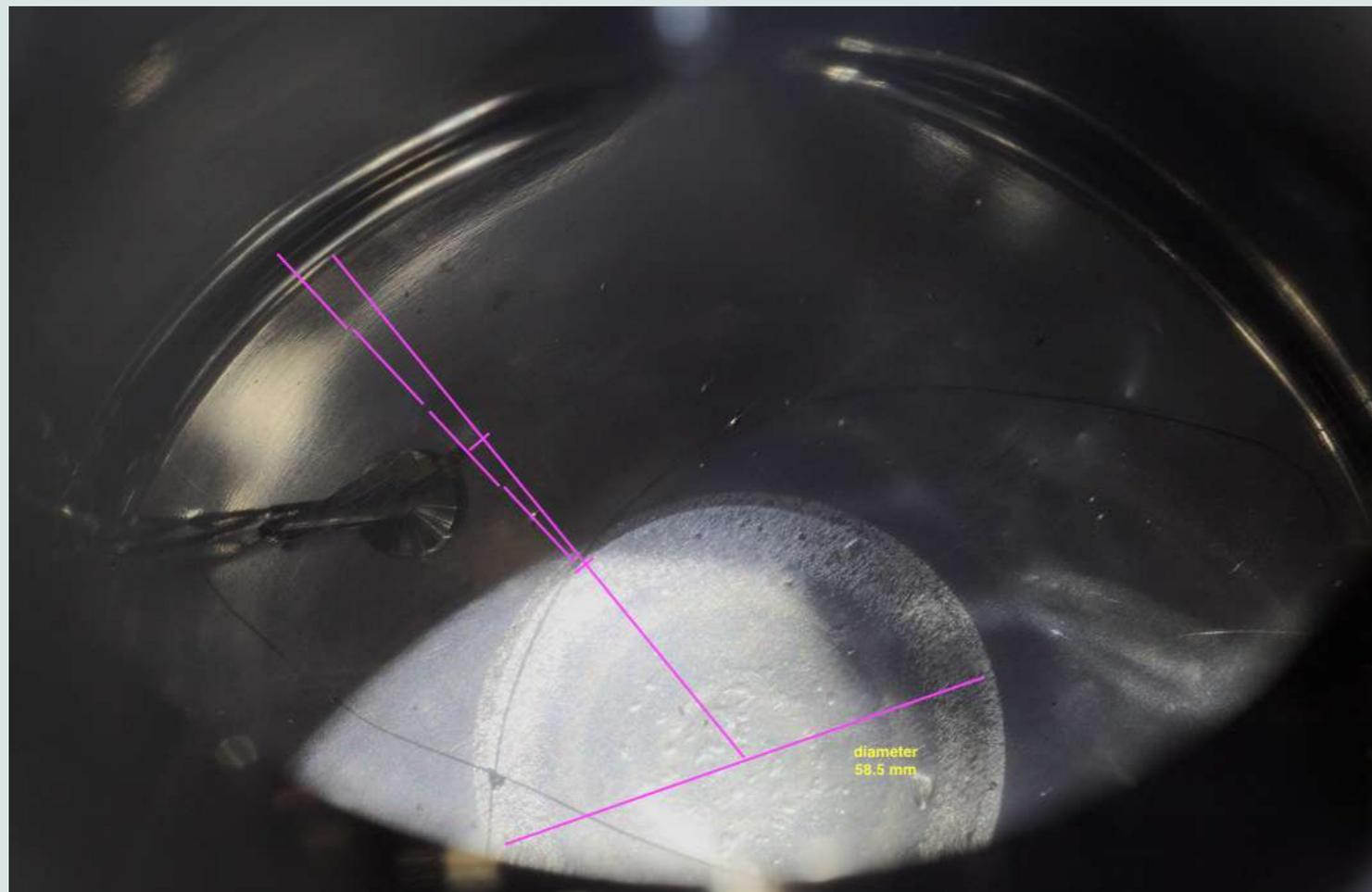
- The main question we want to answer to is what is temperature dependence of the crystal quality?
- We can not measure temperature distribution of the inner wall of SD2 container
- It possible to simulate it using our measured temperatures and reconstructed SD2 shapes



# How we can extract crystal shape from our data



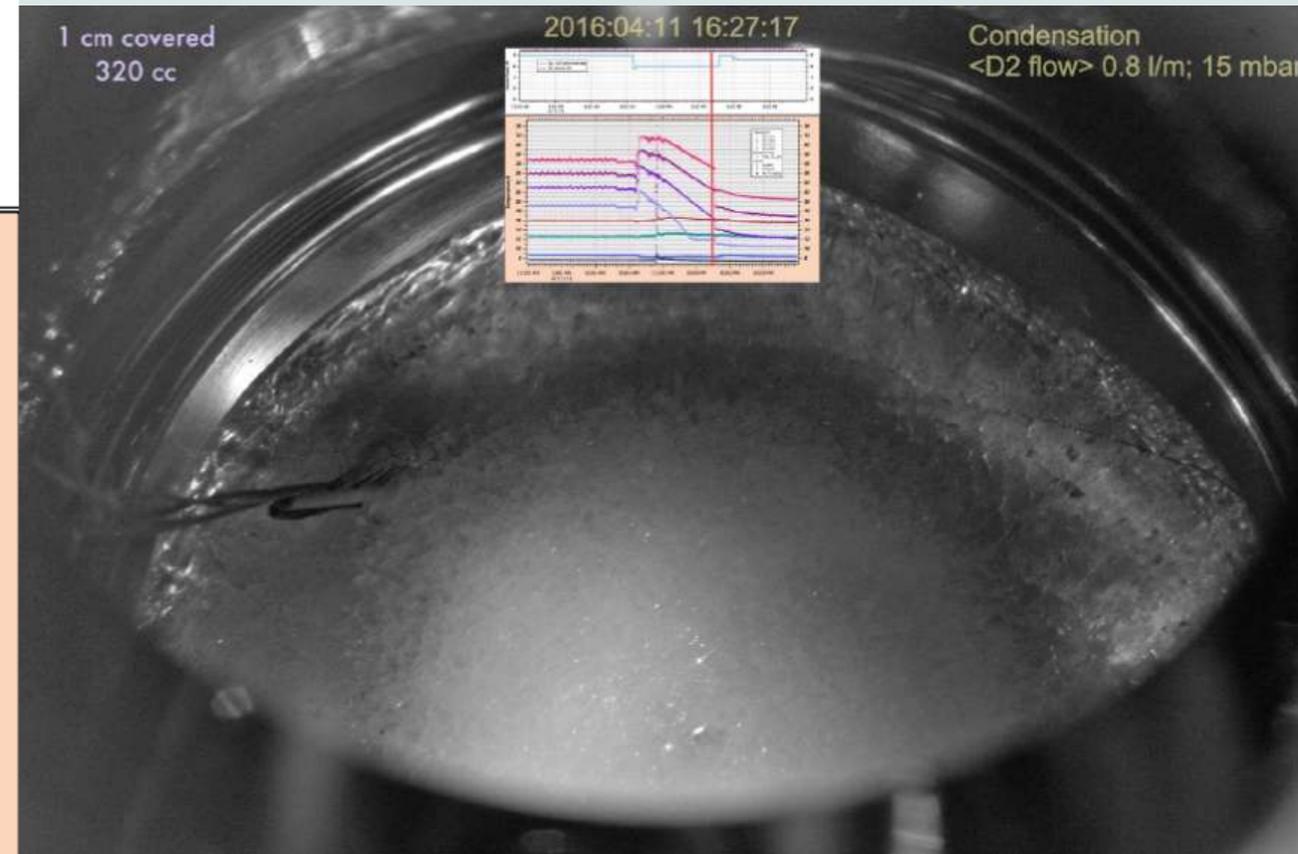
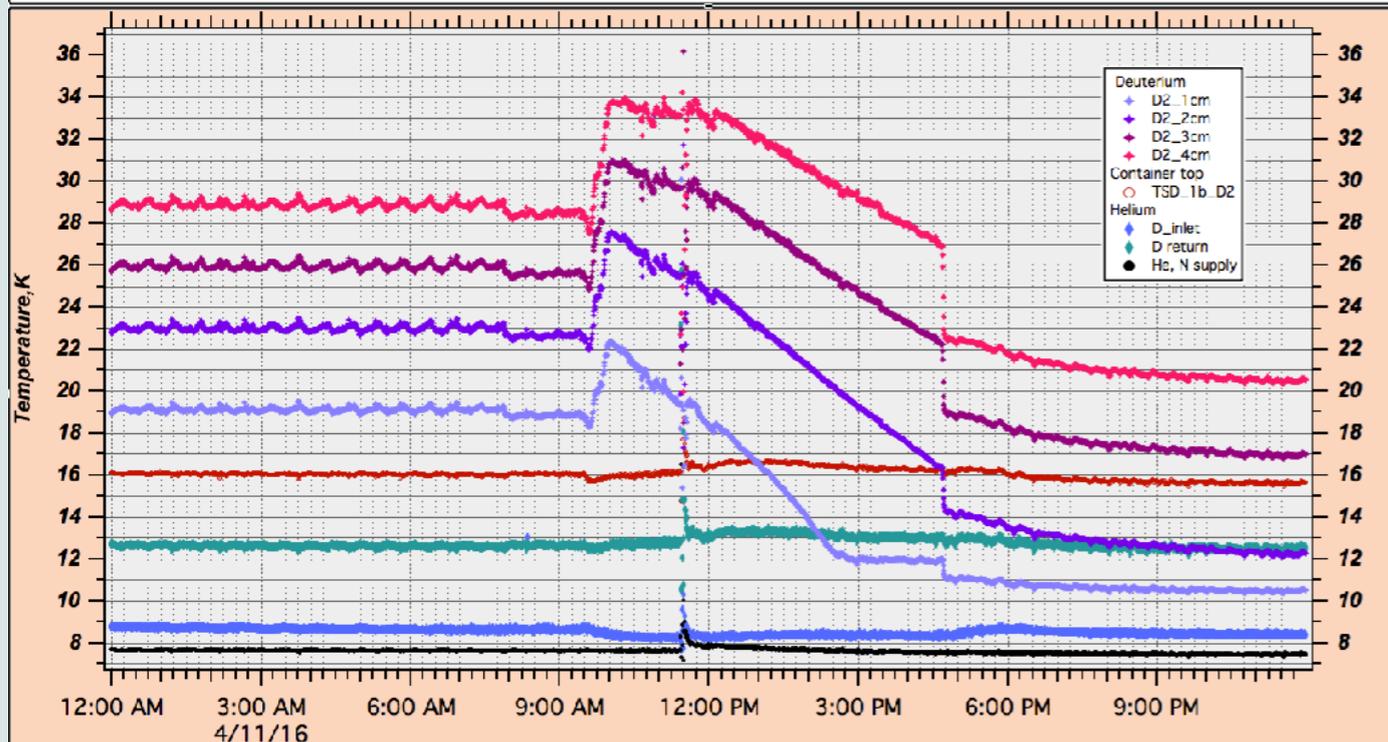
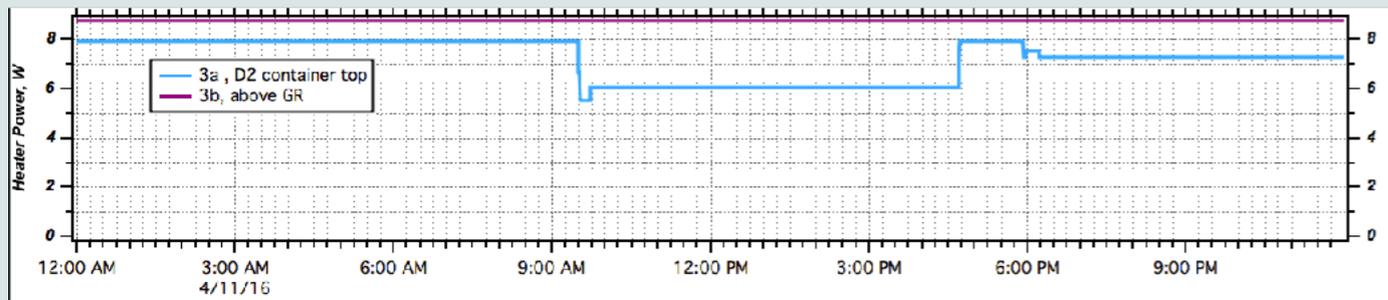
- Position of the T- probe



# How we can extract crystal shape from our data



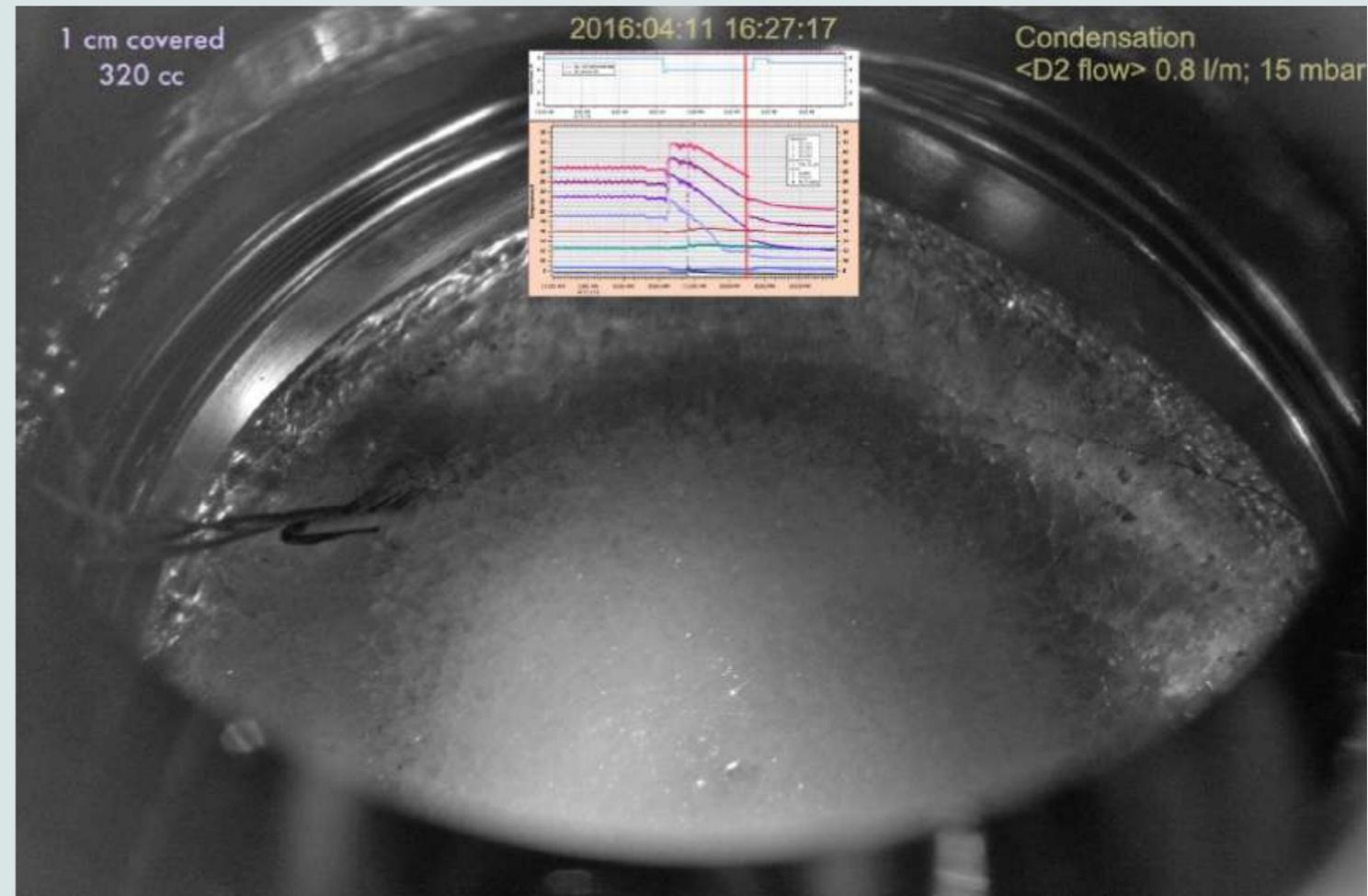
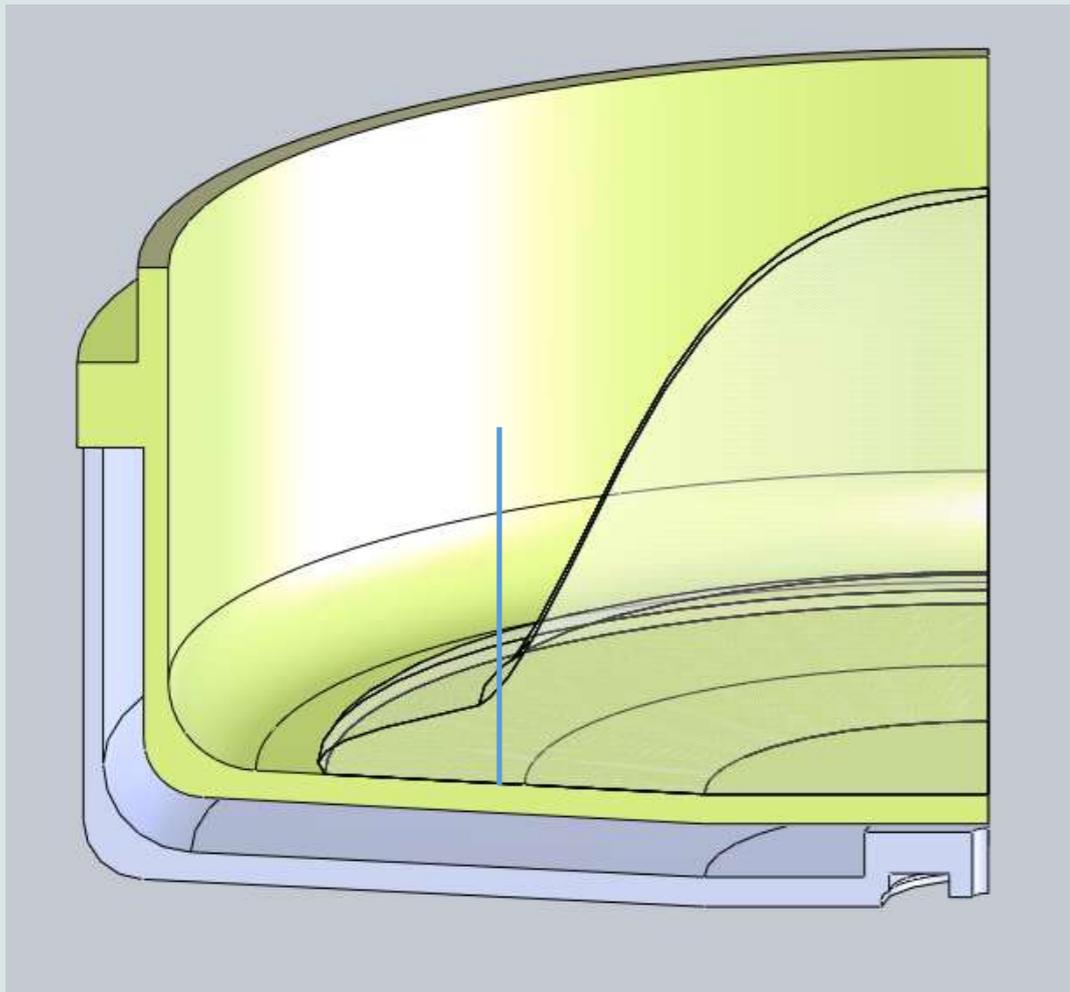
- Hight at probe: coverage of probe sensors



# How we can extract crystal shape from our data



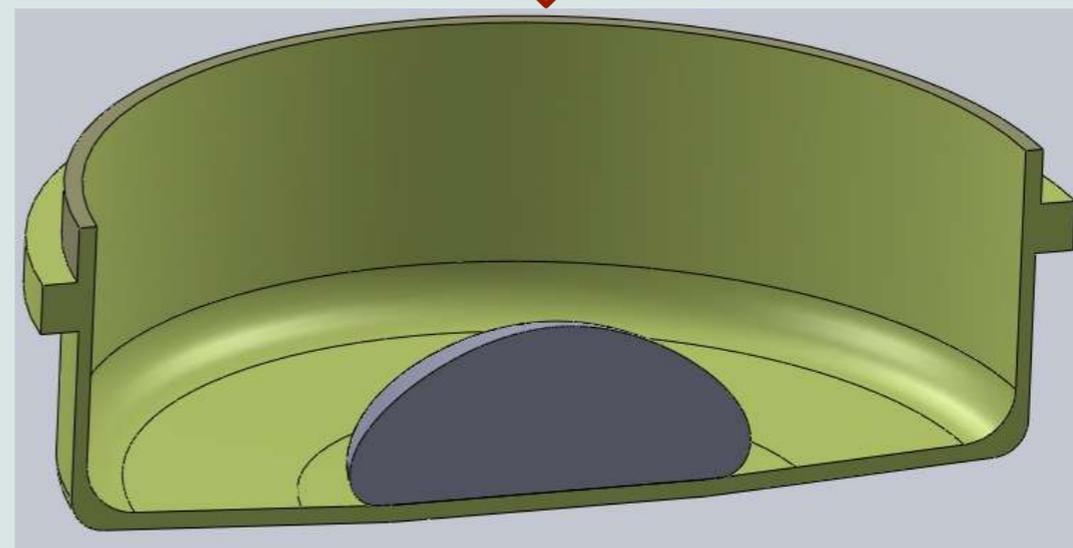
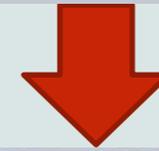
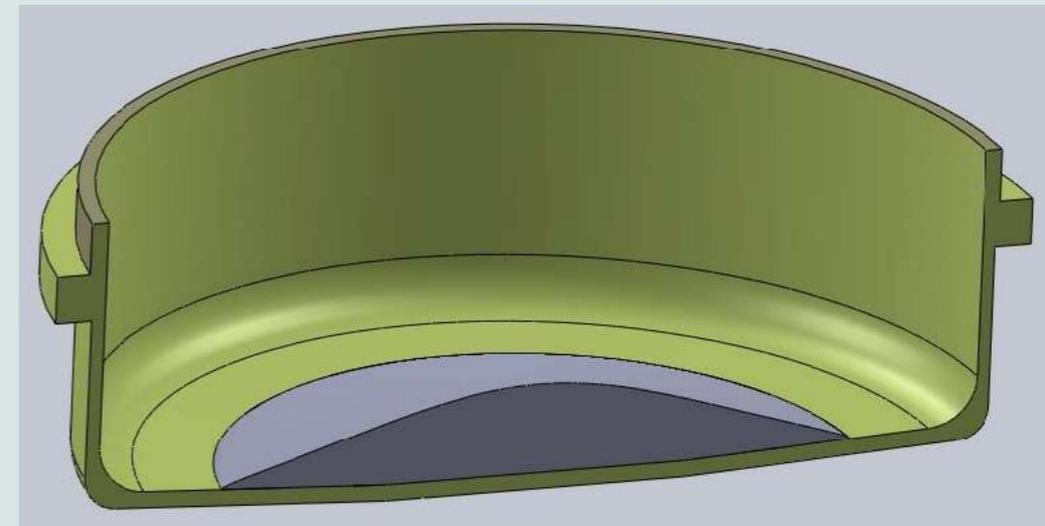
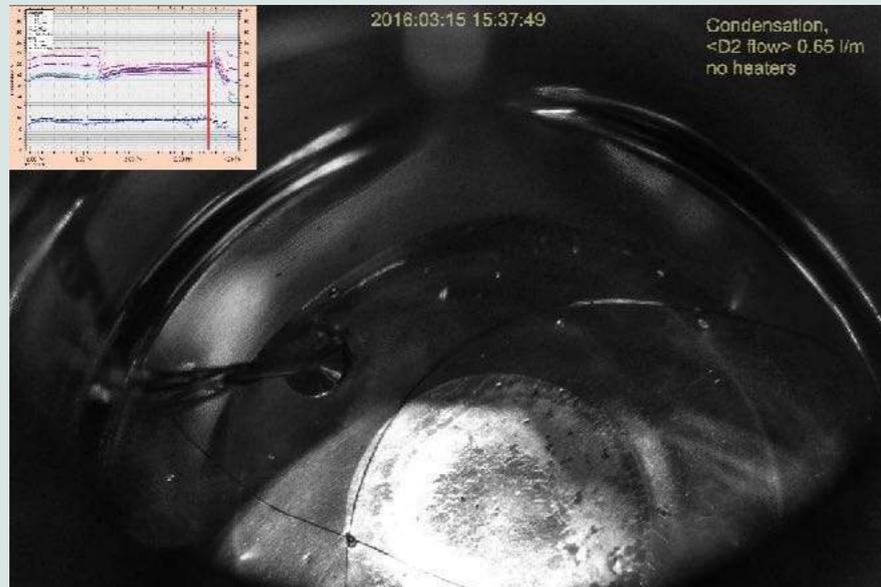
- Volume
- outer diameter of the crystal



# Shape reconstruction



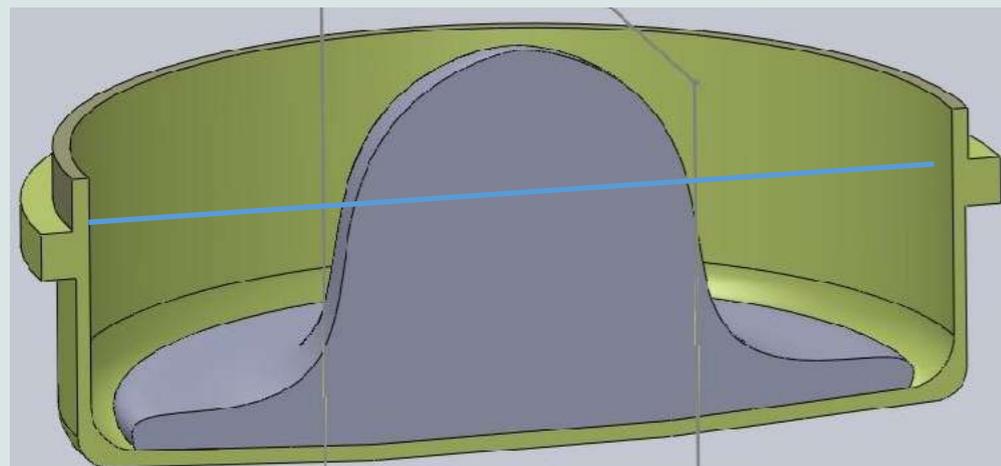
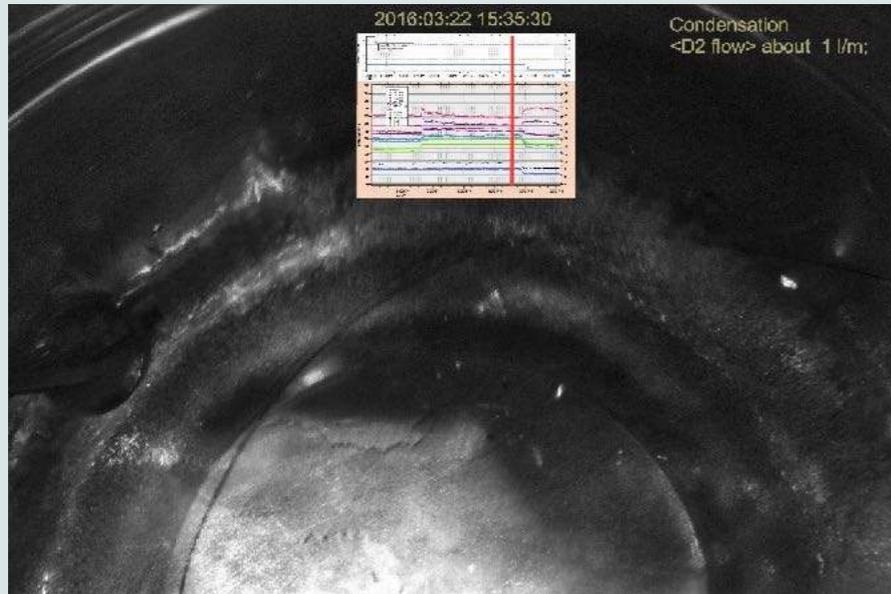
Mar 15, Annealed overnight at 12K. High mobility



# Shape reconstruction (about 350cc)

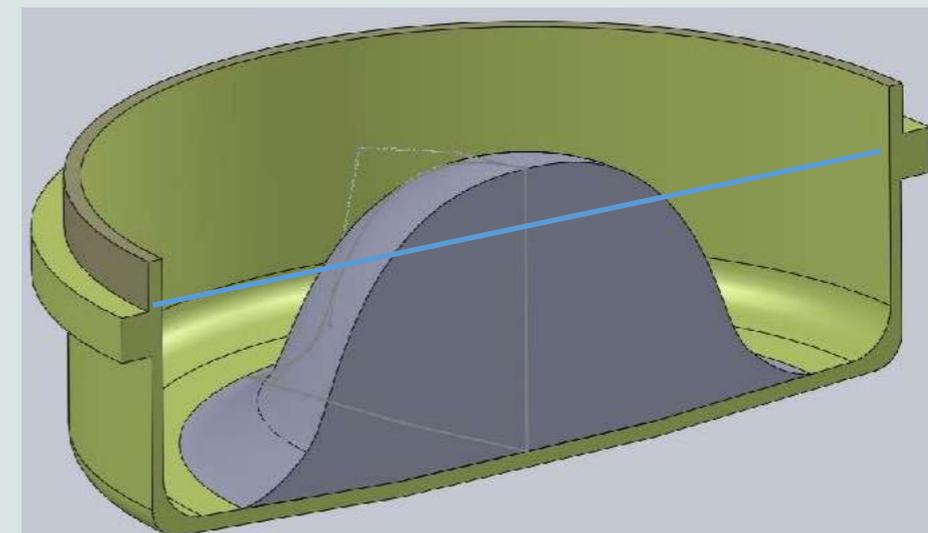
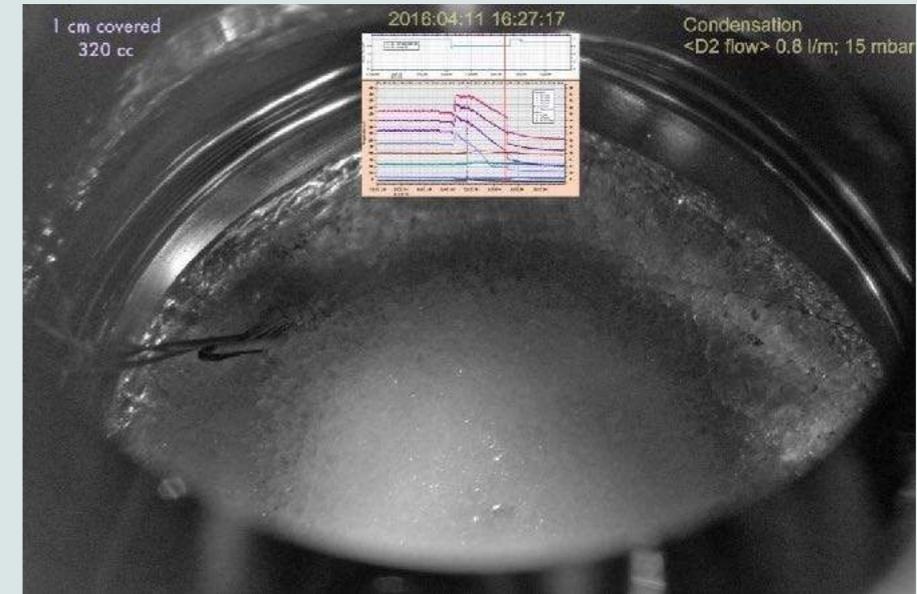


March 22



5 cm

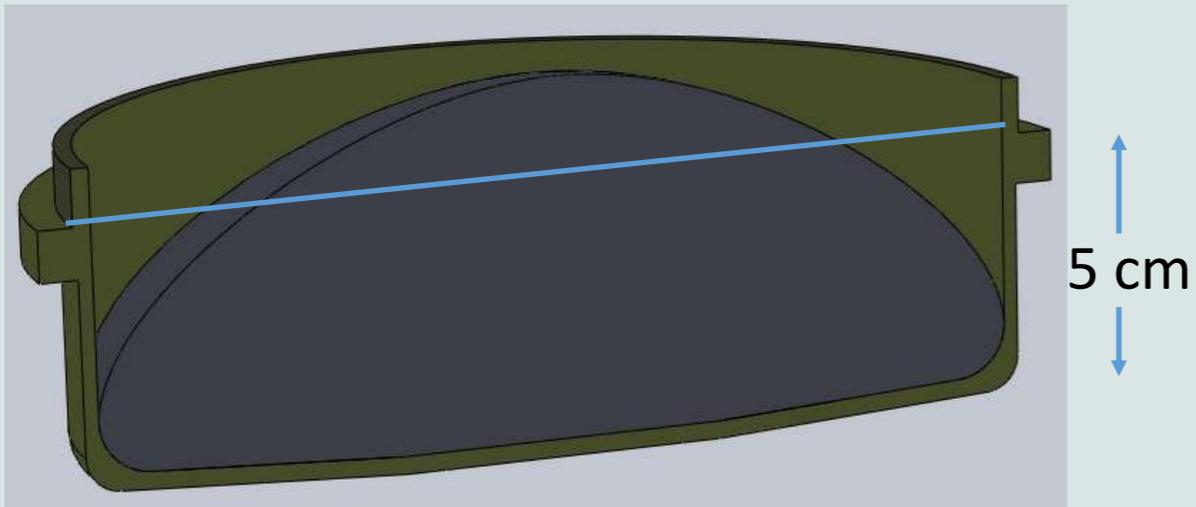
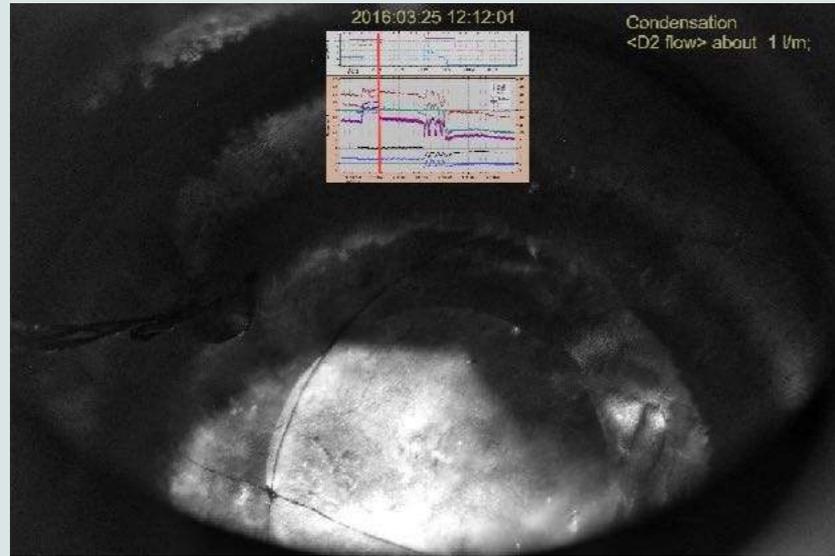
Apr 11



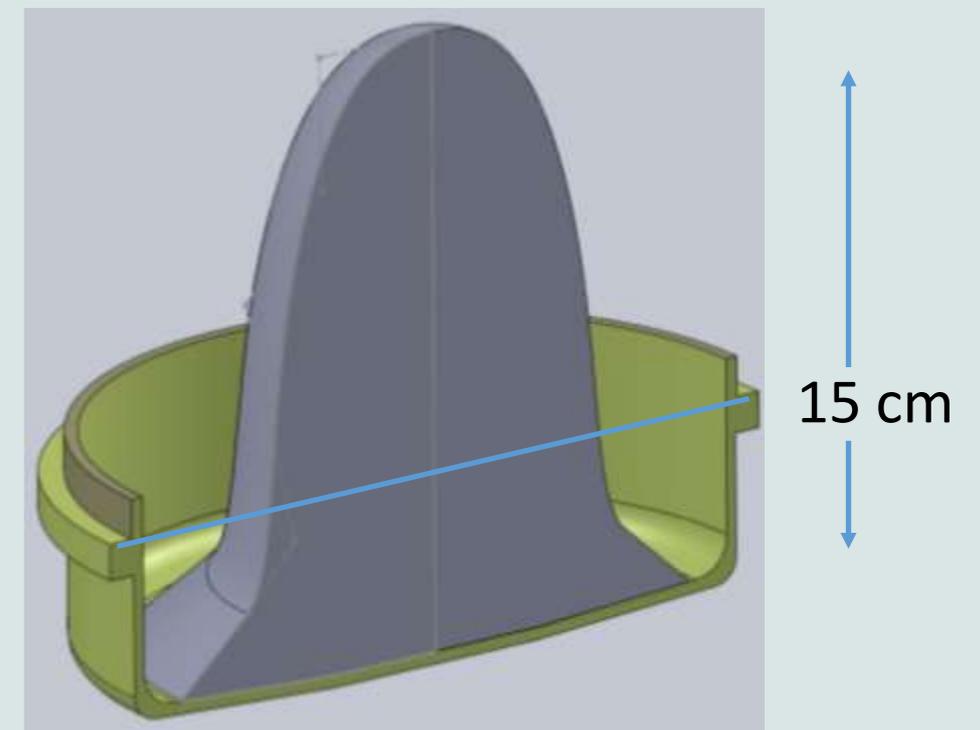
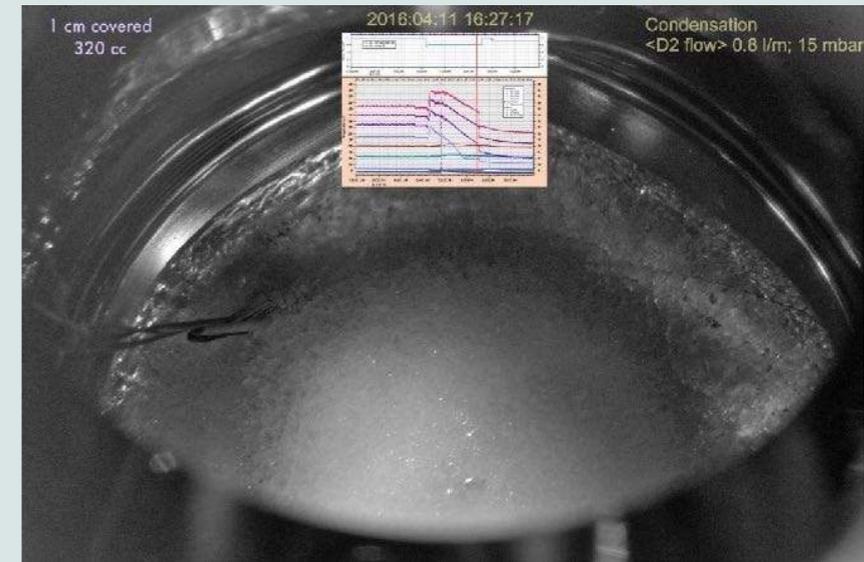
# Shape reconstruction (1050cc vs 964cc)



March 25



April 13



# Conclusion from SD2 shape reconstruction



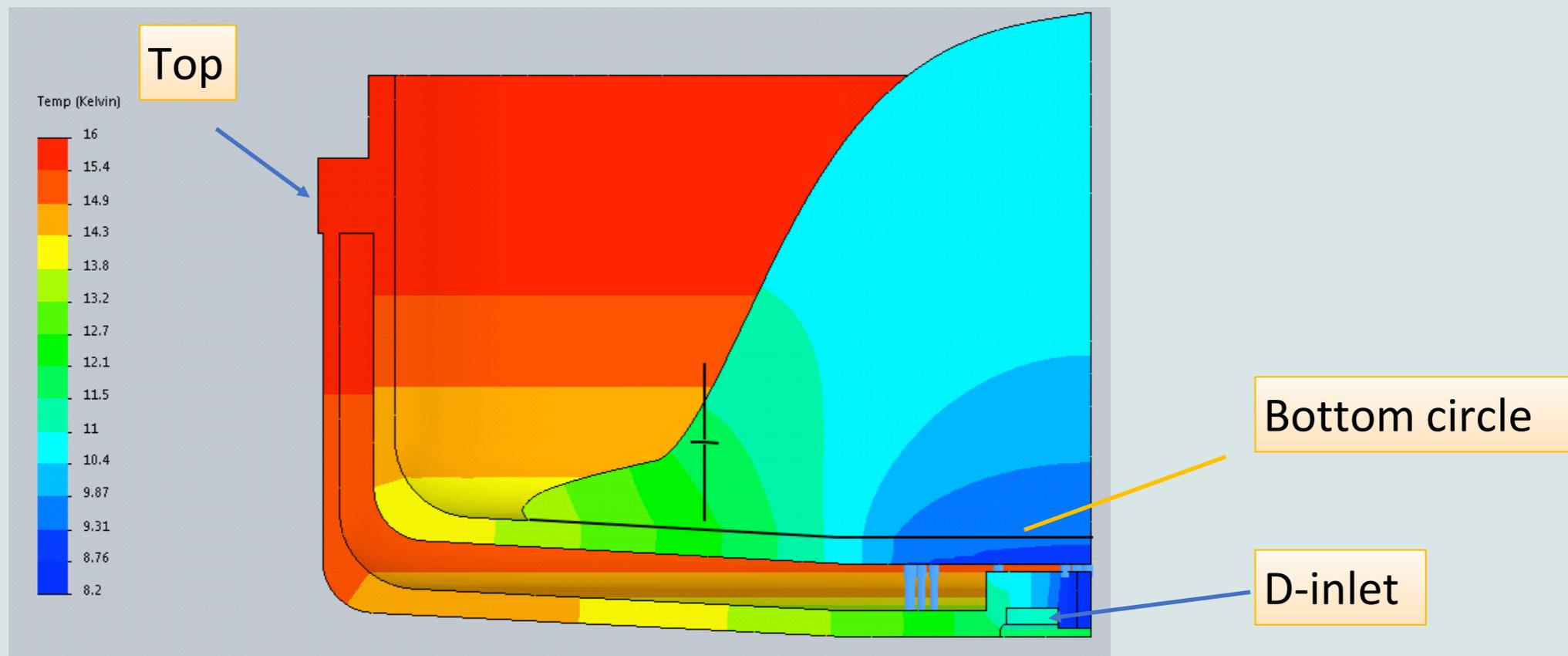
The only difference between March and Apr 11 runs was temperature distribution of the container bottom

Crystal shapes and transparency are extremely temperature dependent

# How to simulate T-distribution of container walls



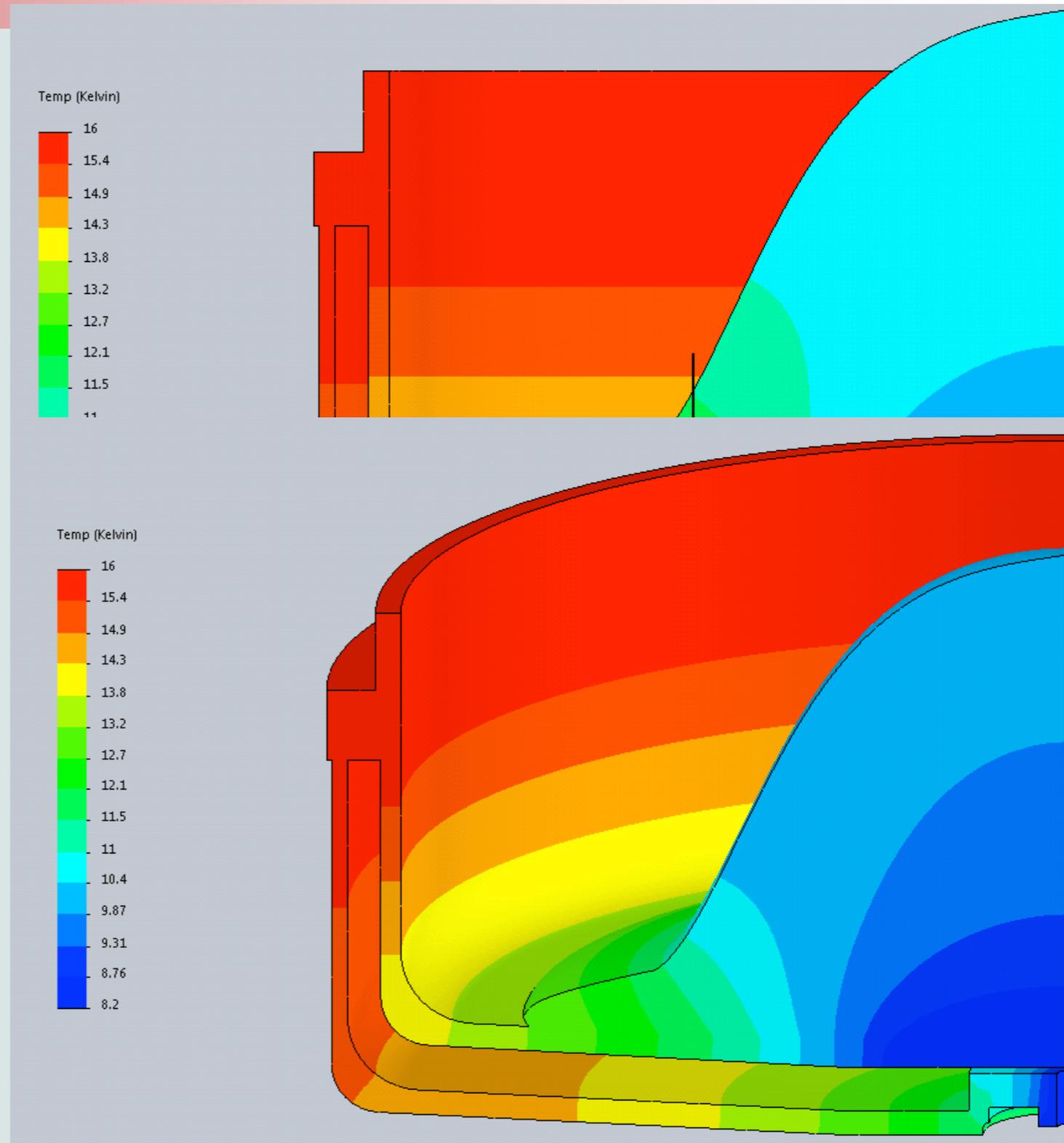
- T-top and T -inlet are fixed on surfaces shown on the left
- Then temperature of the bottom circle is raised above D-inlet until 1 cm reading matches experimental value



# April 11 simulation, D2 flow on and off



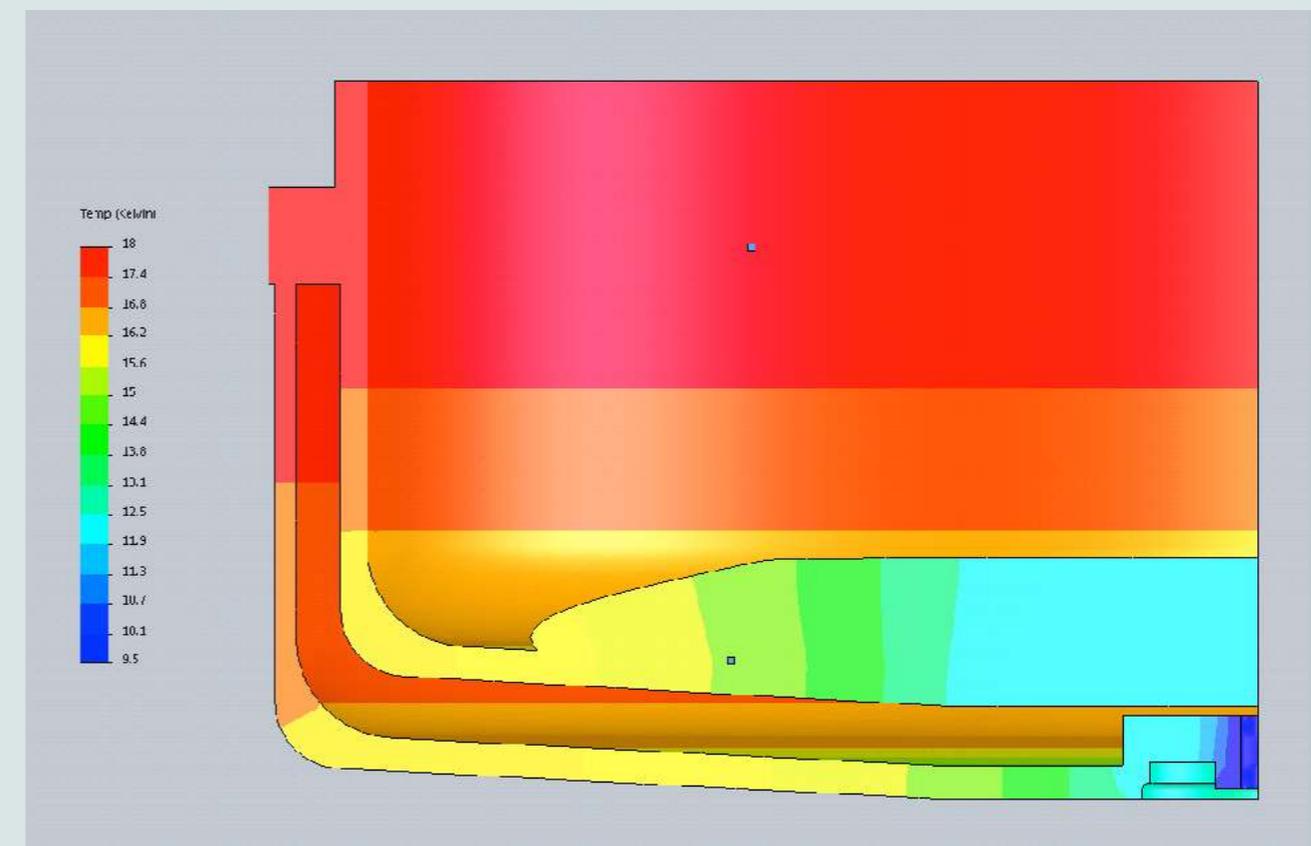
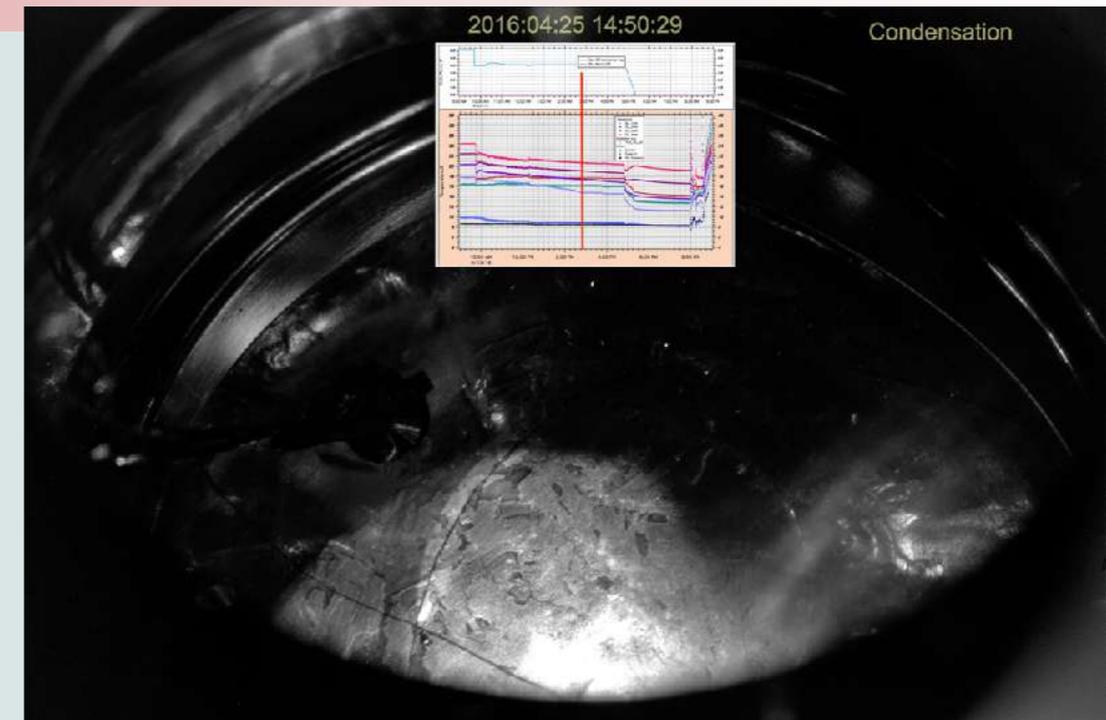
- 1 cm temperature can be reproduced only when assuming that bottom circle is at 9.3K
- Transparent region start from above 12K
- after D2 flow was off, 1 cm dropped to 11K, while the container has not changed, it consistent with Bottom circle at the same T as D-inlet



April 25, 9.5/ 18/ 16.2, 1cm=14.5K, about 340cc total



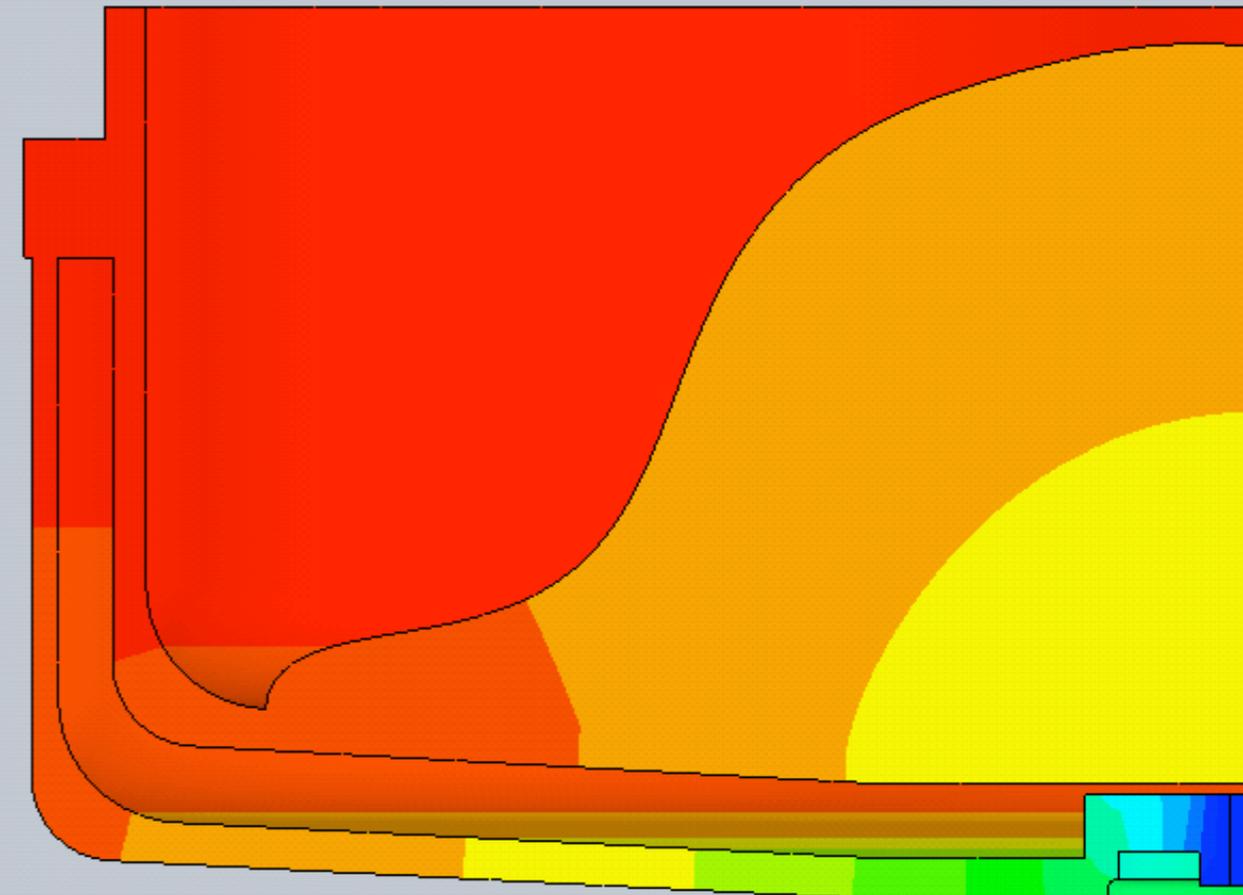
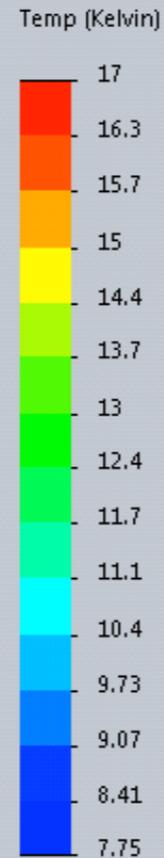
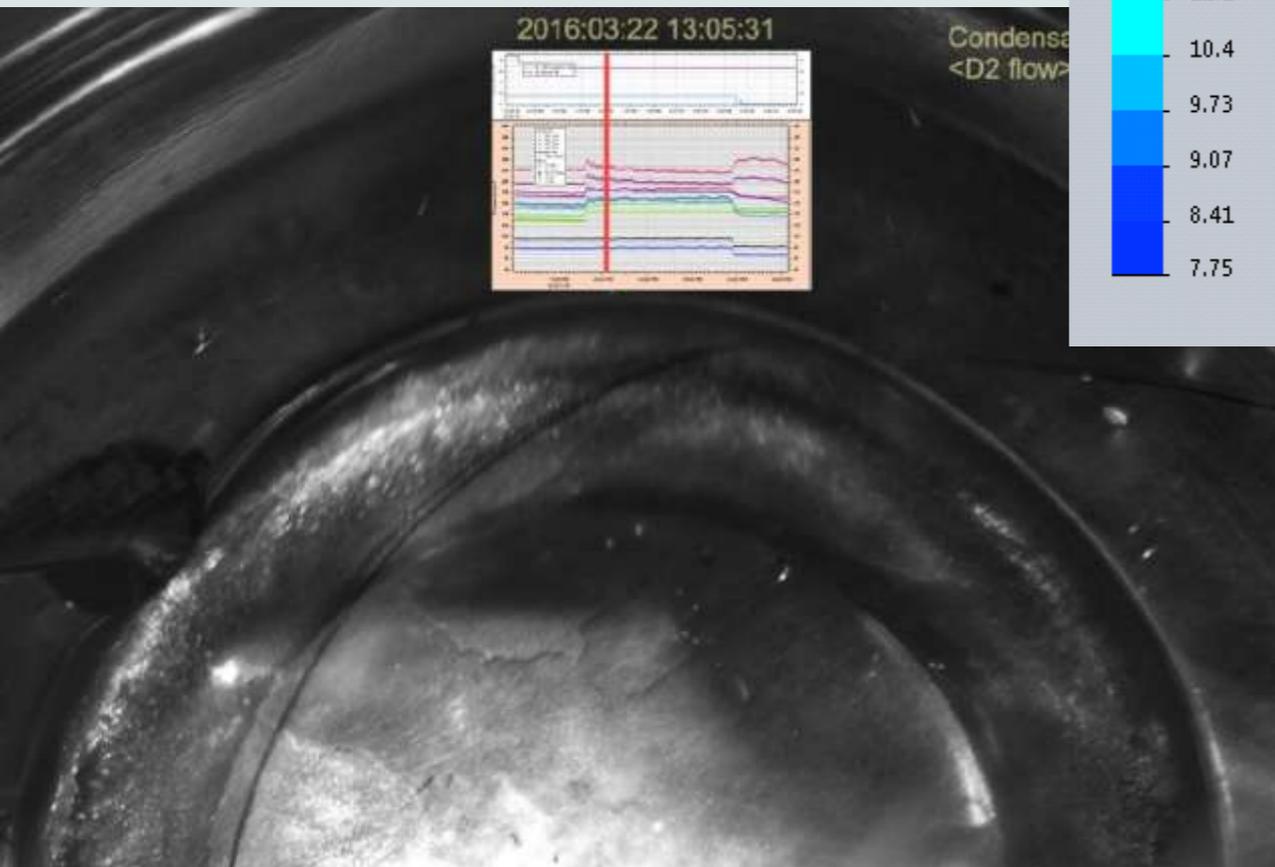
- 1cm temperature can be reproduced only when assuming that bottom circle is at 12K
- **Conclusion:** to grow transparent crystal, cryostat walls needs to be above 12K



Mar 22 condensation, 1cm=16K; 7.75/ 17/ 18.5-19K;  
from simulations bottom centre =14K;



- 1cm temperature can be reproduced only when assuming that bottom circle is at least 14K

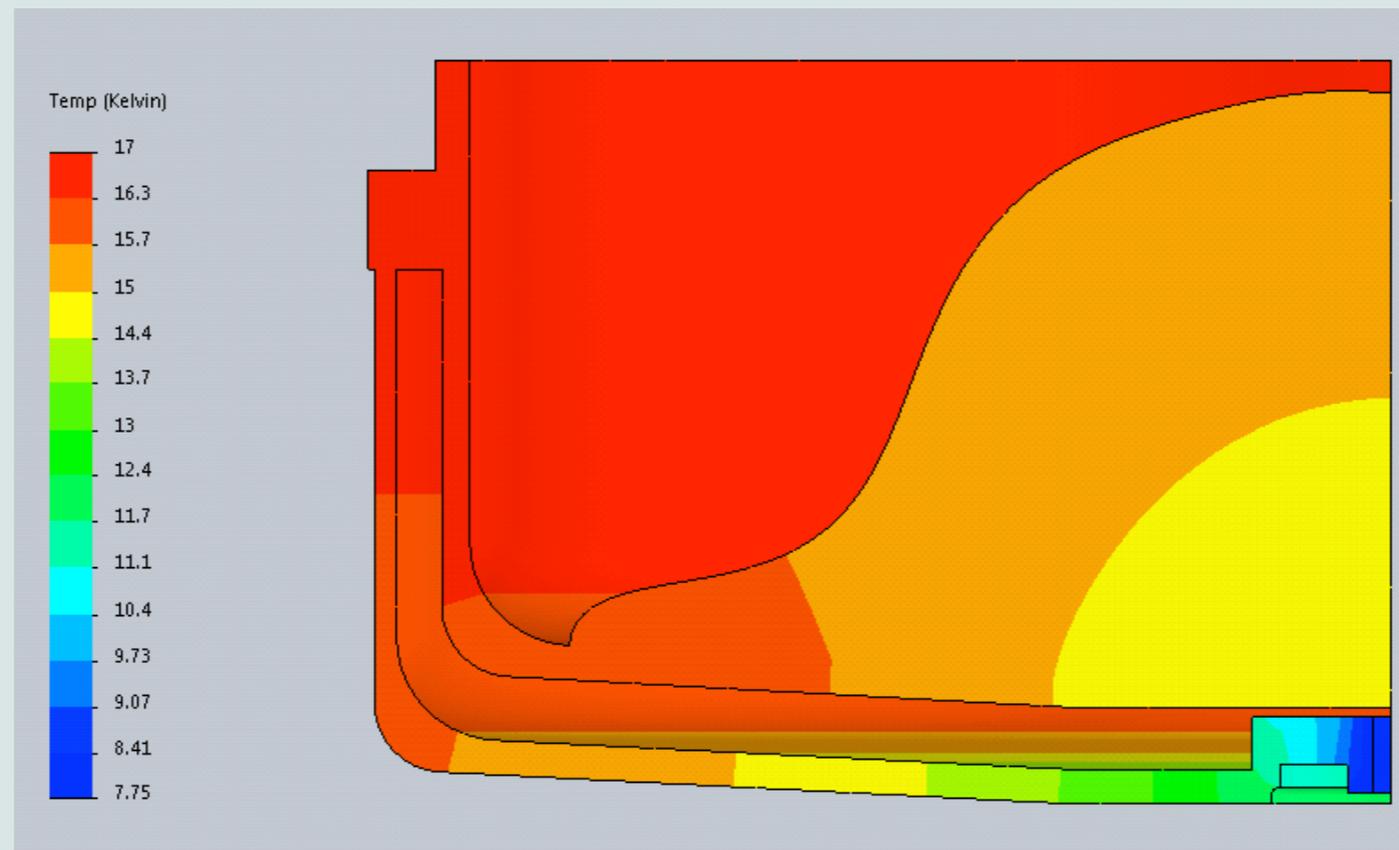


- **Conclusion:** to grow really good transparent crystal, cryostat walls needs to be above 14K

# Conclusion from SD2 Temperature reconstruction



Container walls should be above 12K for growing transparent crystal



# Discussion: MC Simulations of Frost vs Roughness



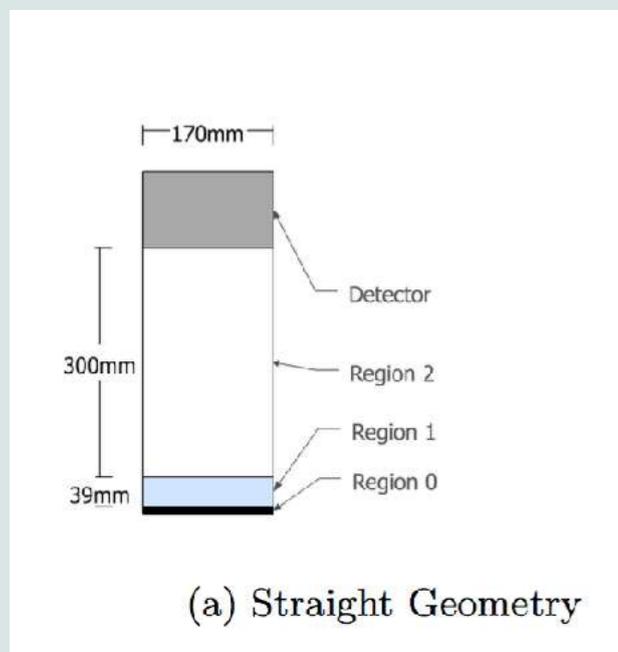
- Transport simulations started with assuming only a rough surface, which wash out completely focusing effect of the SD2 burst
- This model gives transmission reduction about 15%, which is clearly not enough to explain 50% and more observed in real life

# Discussion: MC Simulations of Frost vs Roughness

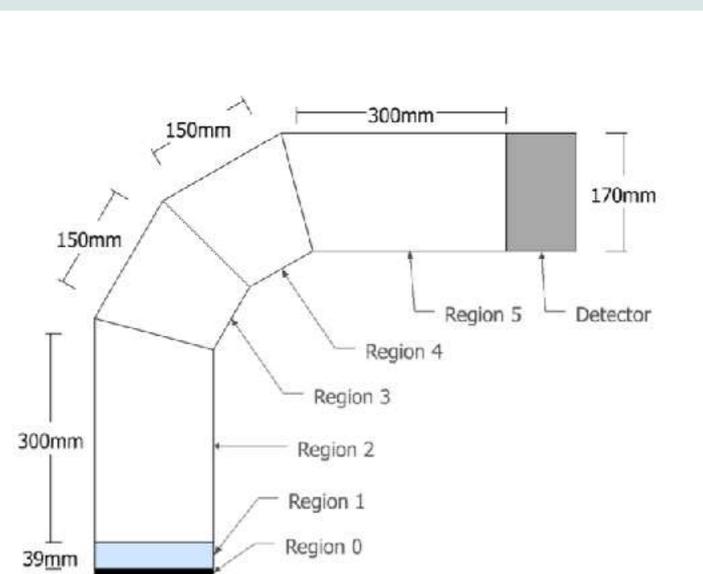


- Frost layers were modeled as SD2 foils parallel to the crystal surface

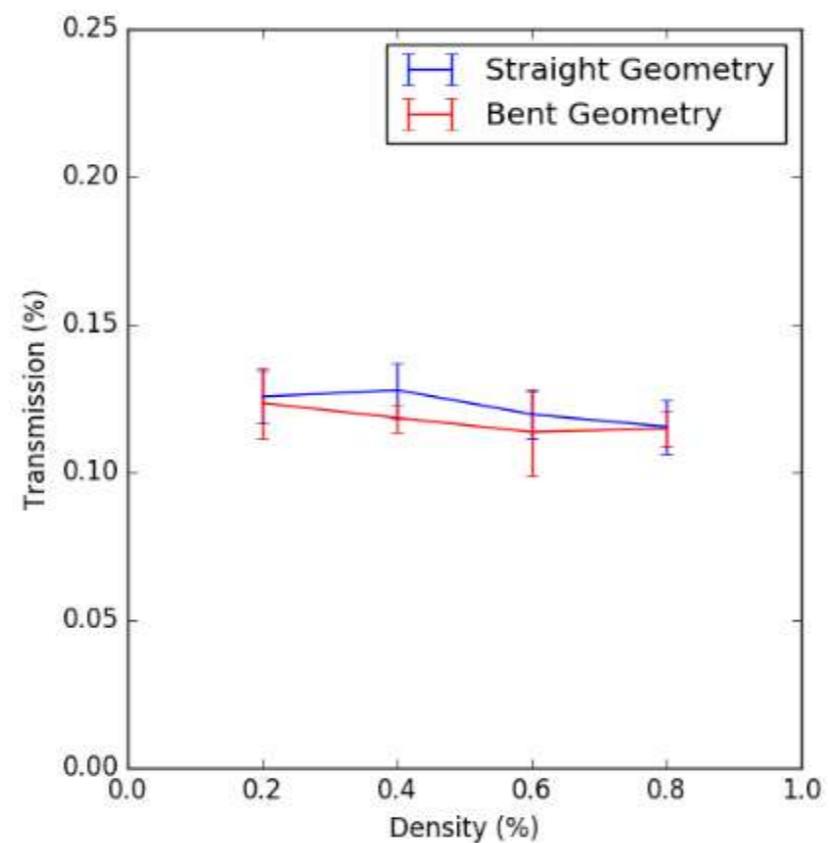
One rough layer can act as a barrier with a decrease in the density of



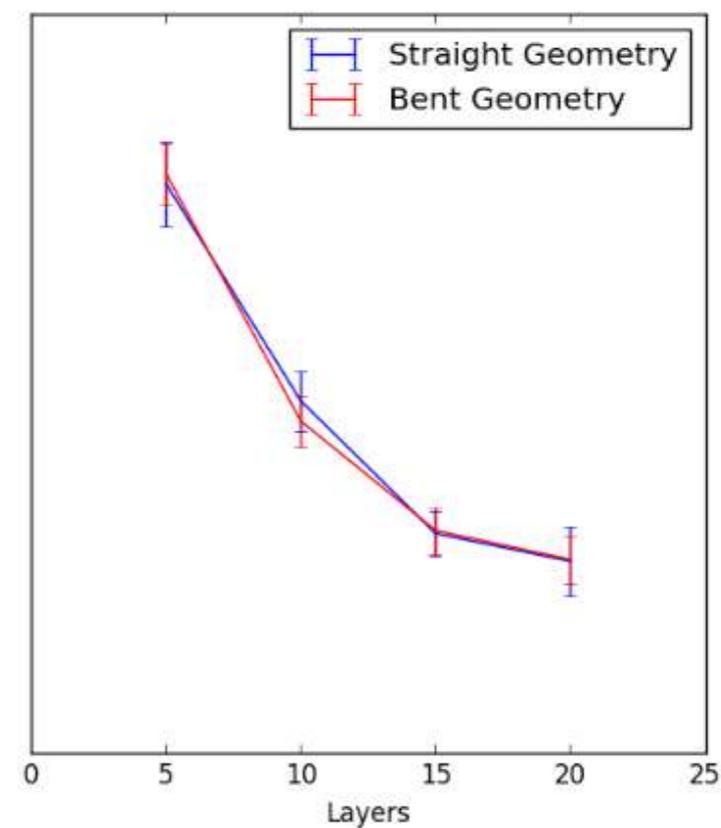
(a) Straight Geometry



(b) Bent Geometry

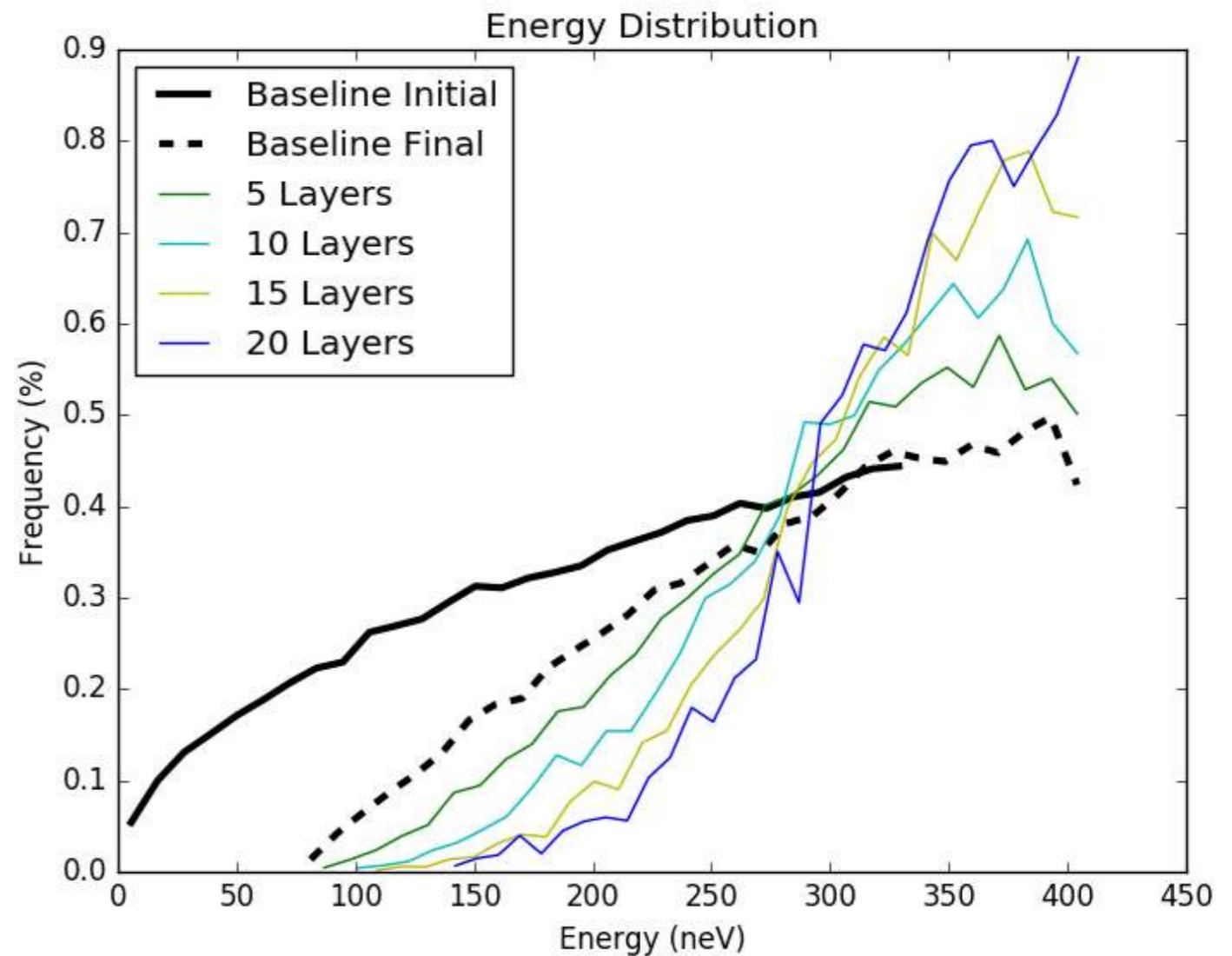
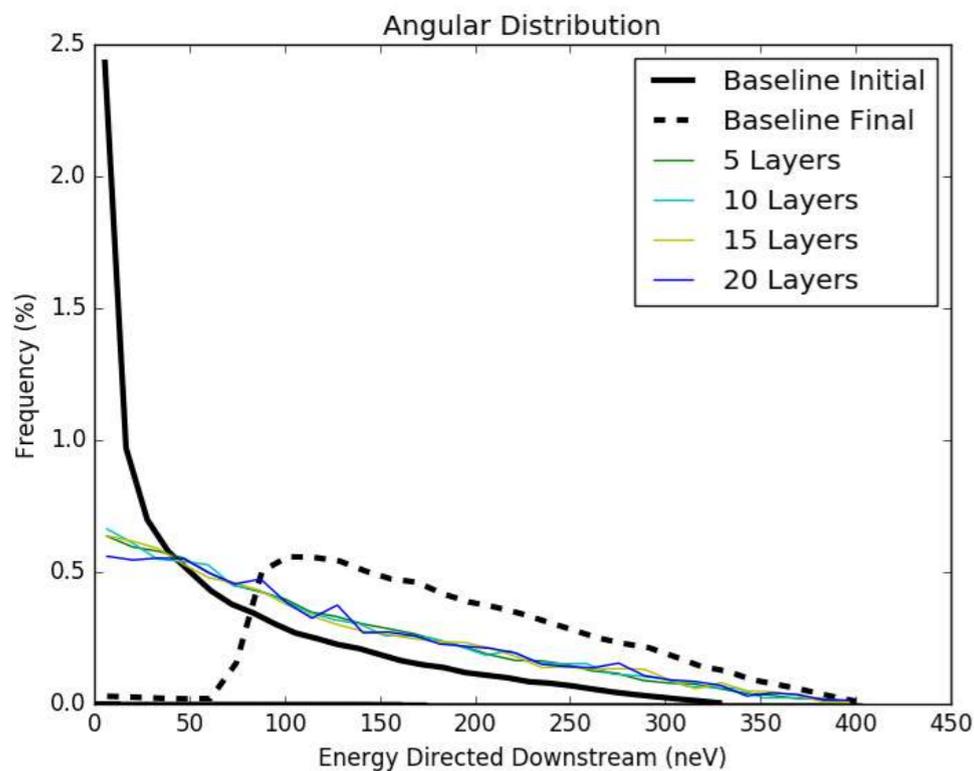
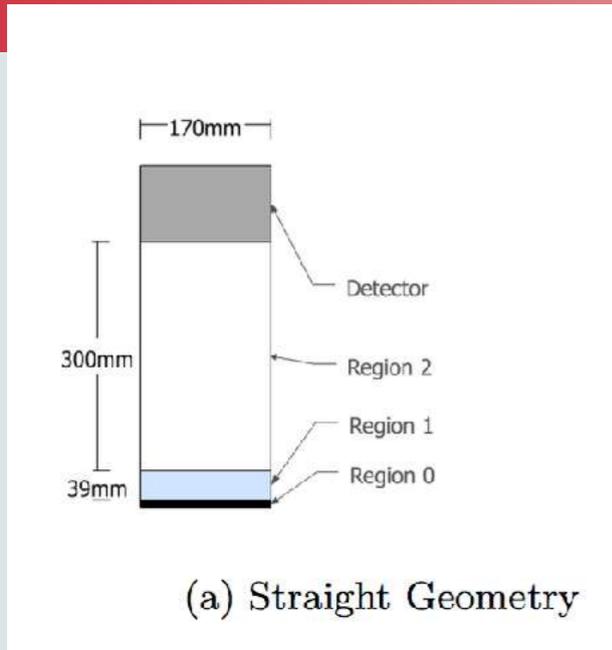


(a) Density

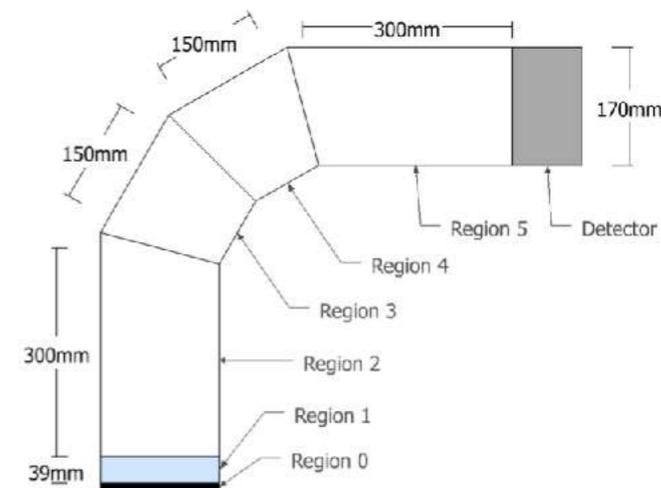


(b) Number of Interactions

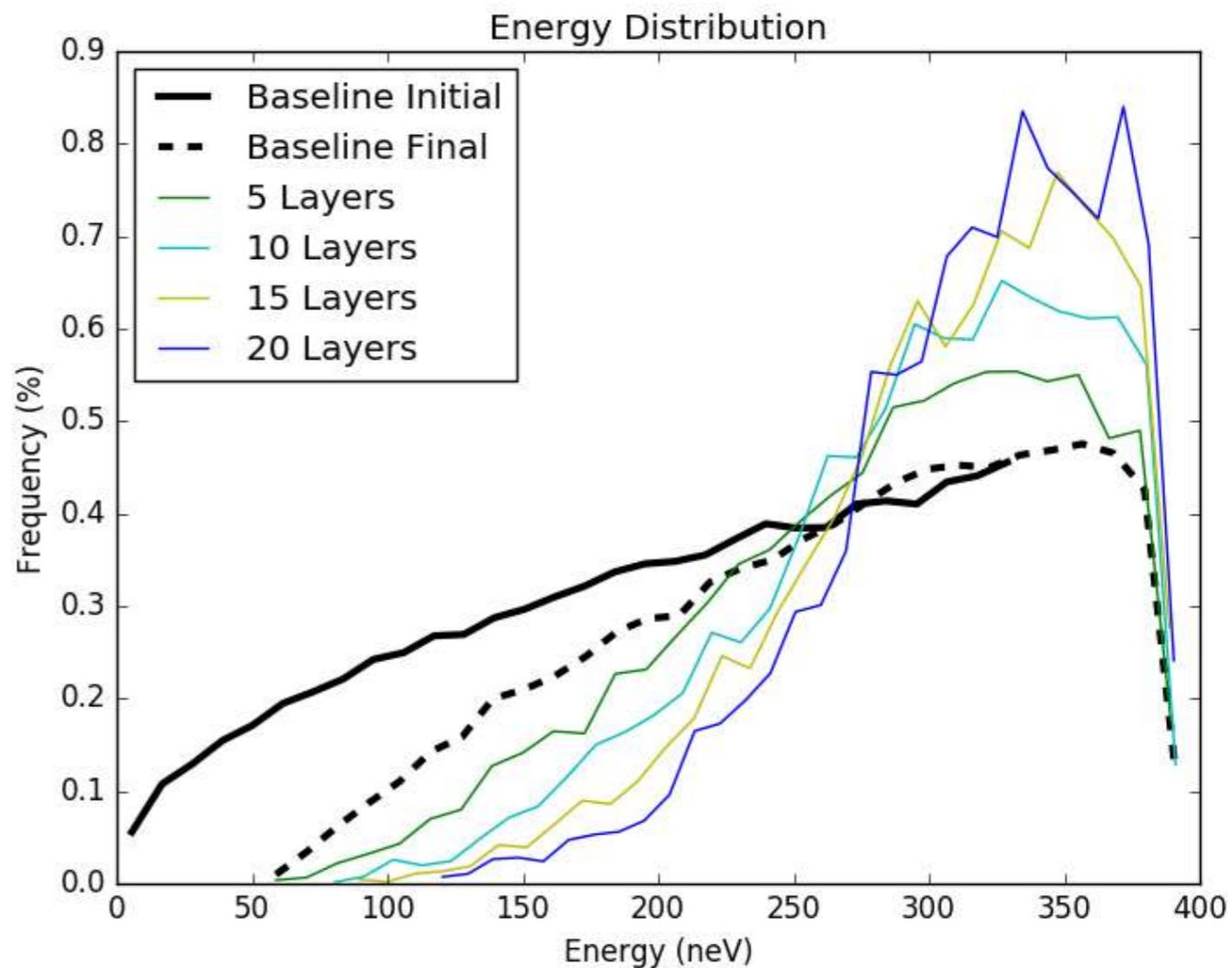
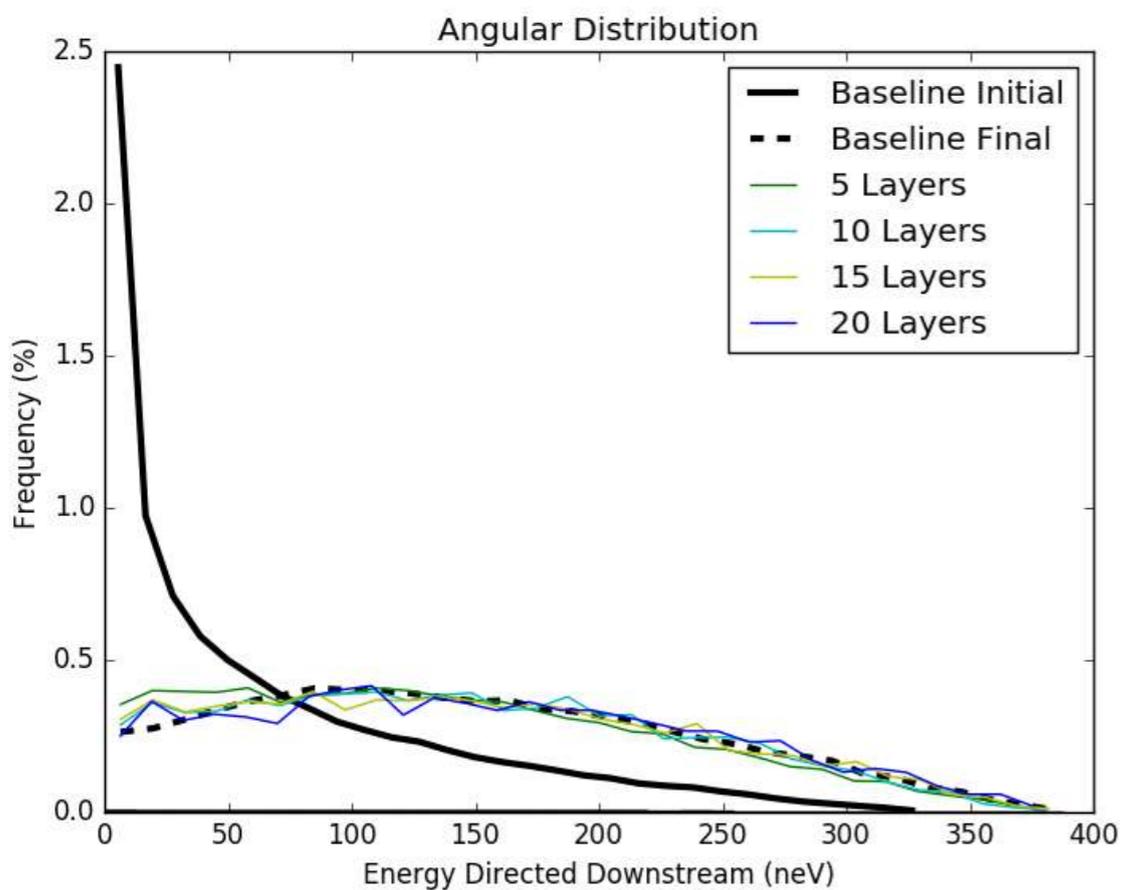
# MC Simulations of the frost layers: Straight



# MC Simulations of the frost layers:



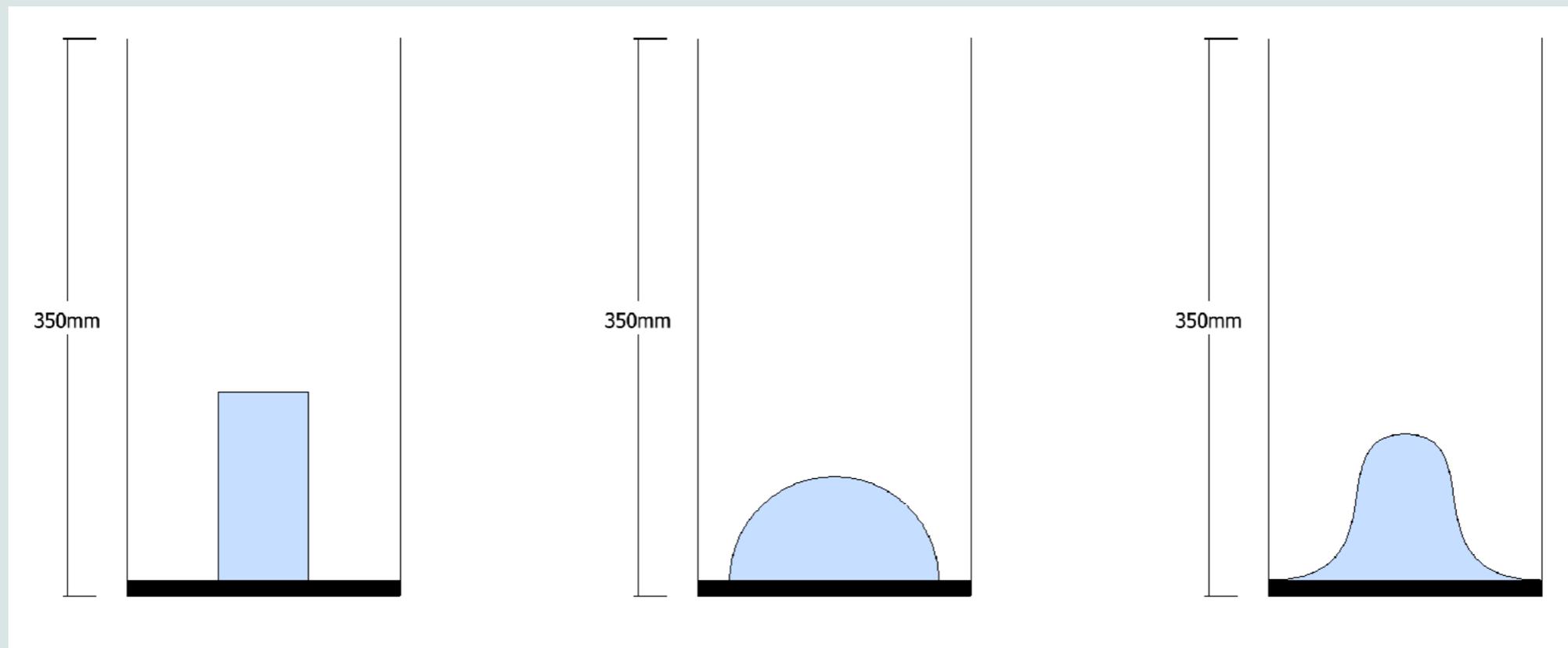
(b) Bent Geometry



# MC Simulations outline: shapes



- The next step would be to simulate effect Transmission vs Crystal Shapes



# Conclusions from our experimental study:



- Both, transparency and shape of the crystal are very temperature dependent
- Not only condensation, but crystal evolution with time needs to be tested in real conditions.
- Strong gradients and temperature oscillations should be avoided
- UCN transport can be affected by different crystal shape and needs to be evaluated

# Outlook



- 2017 :
  - complete manufacturing of the temporary shield
  - another round of SD2 growth
  - install temporary shield
  - start assembling cryostat and external neutron guide on the bio-shield
- 2018 :
  - complete assembling cryostat and external neutron guide on the bio-shield
  - first tests with neutrons on small reactor power

