Solid deuterium and nanodiamonds: the new frontier for Very Cold Neutron production at ESS

Nicola Rizzi

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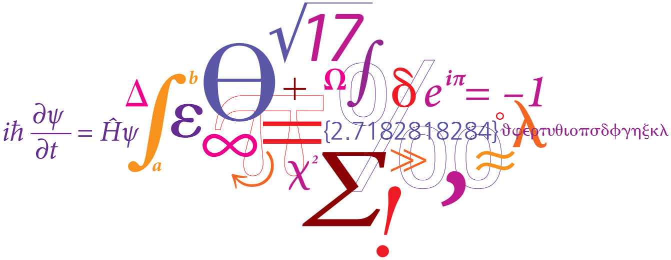
04/2024



Outline

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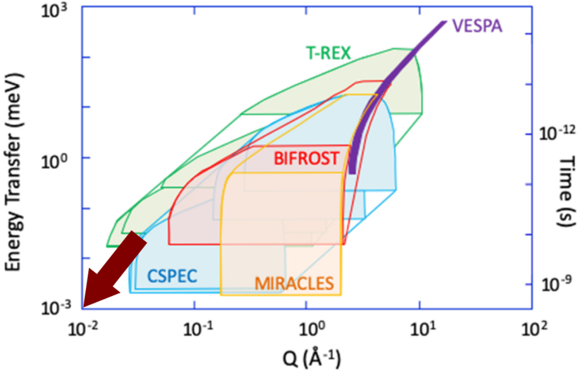
Motivation for this work

- The worldwide trend in neutron research is towards slower neutrons
- There is a problem: neutron sources in this energy range are **weak**!
- It gets worse as energy decreases
- We need more intense sources and a reduction of losses
- Slower neutrons imply longer observation time for fundamental physics and expansion of dynamic range in neutron scattering



The (Q, E) range accessible by the ESS spectrometer suite [1]

In a nutshell

Using validated codes we succeeded in:

Def.	Energy range [meV]	λ-range [Å]	v-range [m/s]
CNs	0.2 to 20	2 to 20	200 to 2000
VCNs	5×10^{-3} to 0.2	20 to 120	30 to 200
UCNs	$< 3 \times 10^{-4}$	> 500	< 8

Introduction

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Using validated codes we succeeded in:

- modeling advanced reflectors for slow neutrons, expanding the capabilities of NCrystal with a plugin

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In a nutshell

HighNess DTU

Using validated codes we succeeded in:

- modeling advanced reflectors for slow neutrons, expanding the capabilities of NCrystal with a plugin
- investigating and optimizing novel designs for a dedicated very cold neutron (VCN) source to be built at the European Spallation Source (ESS)
- identifying the best in-pile designs for an Ultra-cold Neutron (UCN) source at ESS
- designing a mock-up experiment to characterize the reflectivity properties of advanced materials

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NCrystal Thermal Neutron Transport

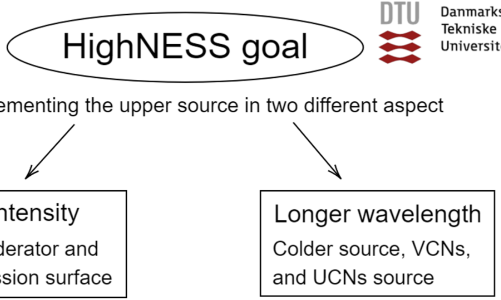
MCNP MONTE CARLO N-PARTICLE

McStas n

DAKOTA Explore and predict with confidence.

HighNESS

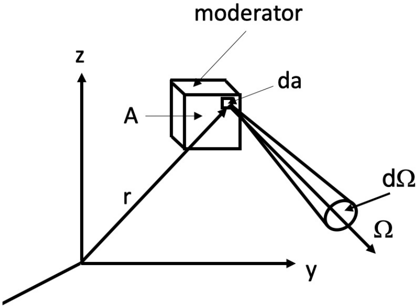
High Intensity **N**eutron Source at the **E**uropean **S**pallation **S**ource



HighNESS is funded by the European Union Framework Programme for Research and Innovation Horizon 2020, under grant agreement 951782

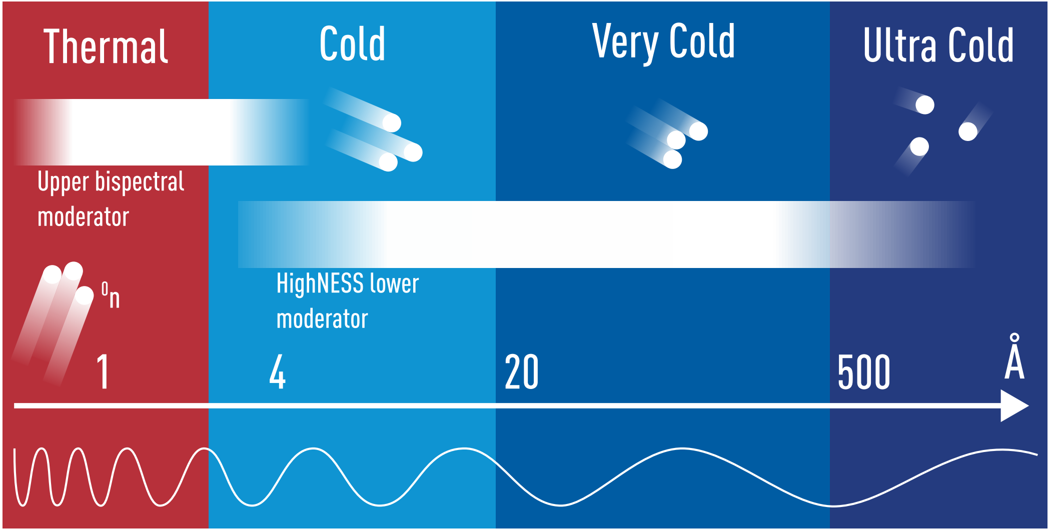
Intensity

- Brightness integrated over the moderator surface
- Prioritizes the total number of neutrons over beam divergence

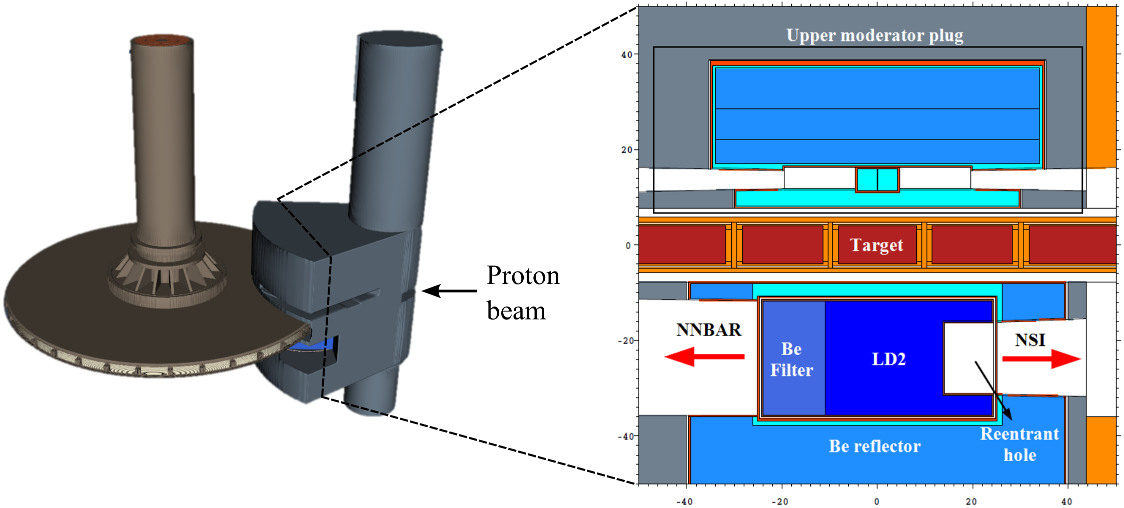


$$I_N(\vec{\Omega}, E, t) = \int_A da\, \Phi(\vec{r}, \vec{\Omega}, E, t) \quad [\mathrm{n/s/sr/ev}]$$

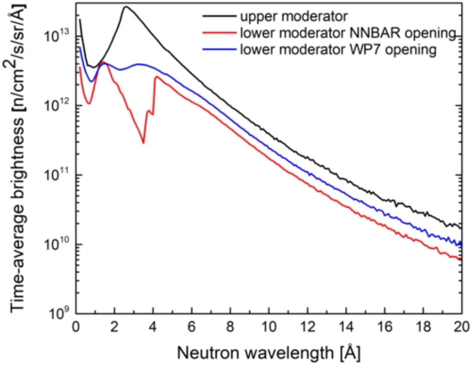
Energy shift

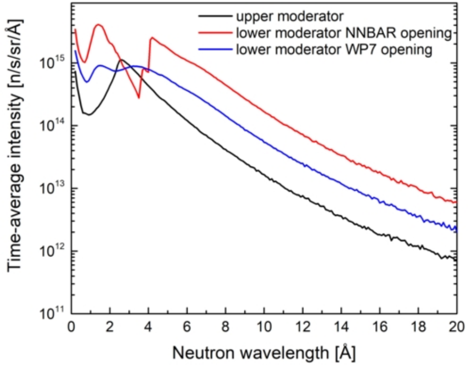


The cold moderator



Upper vs Lower moderator





Ref.[8]

VCN options for ESS

As part of the HighNESS project, this thesis aimed to study and optimize the options for a VCN source at ESS. In the original proposal:

1. **Advanced reflectors** to selectively enhance the VCN transport from the cold source
2. Dedicated in-pile VCN moderator

VCN options for ESS

As part of the HighNESS project, this thesis aimed to study and optimize the options for a VCN source at ESS. In the original proposal:

1. **Advanced reflectors** to selectively enhance the VCN transport from the cold source
2. Dedicated in-pile VCN moderator

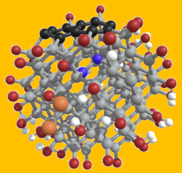
Recently, Valery Neshvizevsky proposed a third concept:

3. Combined use of a cold source, **solid ortho-deuterium** (SD_2) and **advanced reflectors**

Advanced reflectors

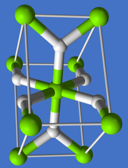
Nanodiamonds

- VCN reflector
- Thermal scattering library developed within HighNESS



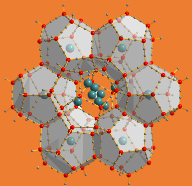
MgH_2

- CN and VCN reflector
- Thermal scattering library available



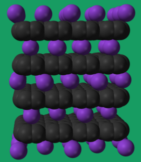
Clathrates hydrates

- Possible VCN converter or VCN reflector
- Cross section measurements and thermal scattering library within HighNESS



GIC

- Possible VCN reflector
- Cross section measurements and thermal scattering library determination within HighNESS



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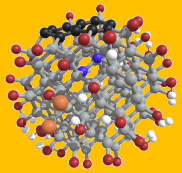

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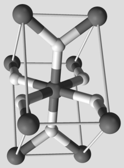

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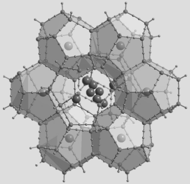

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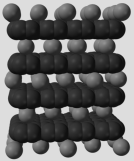

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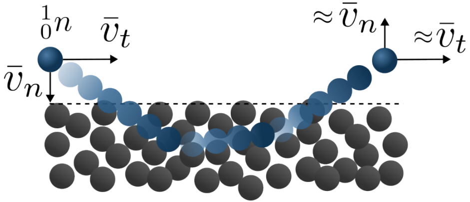
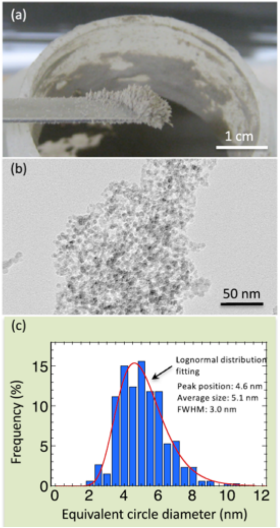
What are nanodiamonds (NDs)?

- A **diamond core** within an onion-like shell measuring few nanometers
- ND powder samples showed efficient reflector properties for very cold neutrons (VCN)
- quasi-specular reflectivity for cold neutrons
- small absorption cross section





Ref.[9]

SANSND NCrystal Plugin

NUCLEAR SCIENCE AND ENGINEERING
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DOI: https://doi.org/10.1080/00295639.2023.2196926

Check for updates

Benchmarking of the NCrystal SANS Plugin for Nanodiamonds

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[b]*European Spallation Source ERIC, Lund, Sweden*
[c]*UGA Graduate School of Engineering in Physics, Electronics, Materials Sciences, Grenoble INP – Phelma, 3 Parvis Louis Neel – CS 50257 – 38016 Grenoble Cedex 1, France*

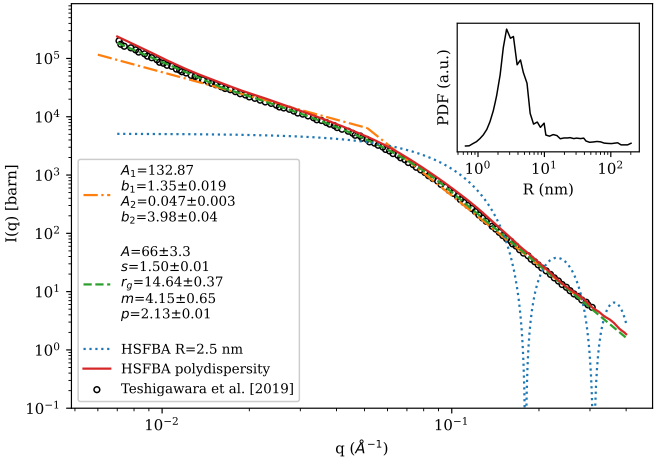
Received November 21, 2022
Accepted for Publication March 25, 2023

Empirical and theoretical models

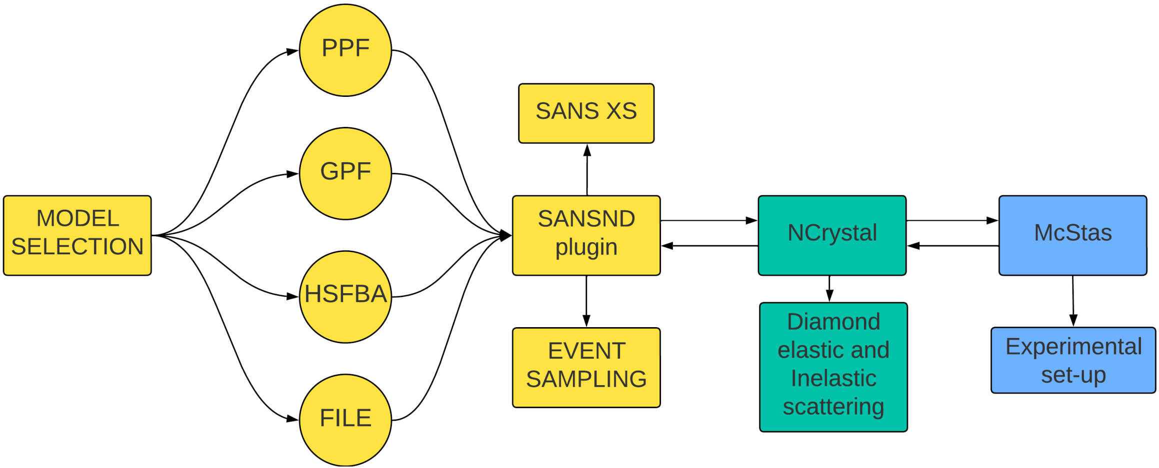
- Empirical models based on experimental $I(q)$ in absolute units:

$$\sigma_{SANS}(k_0) = \frac{2\pi}{k_0^2}\int_0^{2k_0} qI(q)\,\mathrm{d}q$$

- Theoretical model based on hard sphere model to calculate the scattering amplitude per nanoparticle in the first Born approximation.

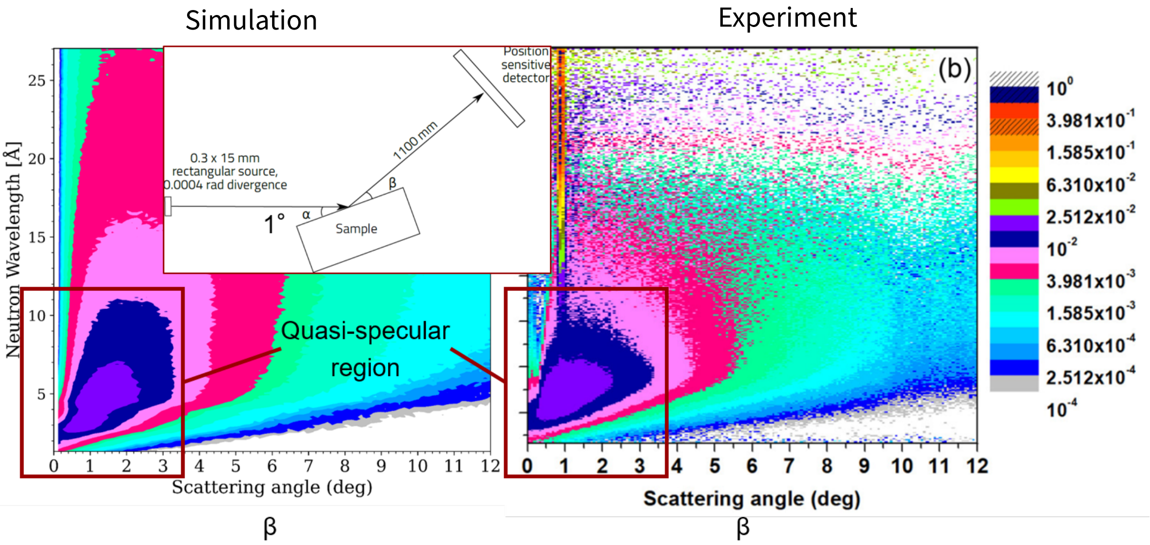


How to use it


MODEL SELECTION
PPF
GPF
HSFBA
FILE
SANS XS
SANSND plugin
EVENT SAMPLING
NCrystal
Diamond elastic and Inelastic scattering
McStas
Experimental set-up

Nanodiamonds

Probability of scattering



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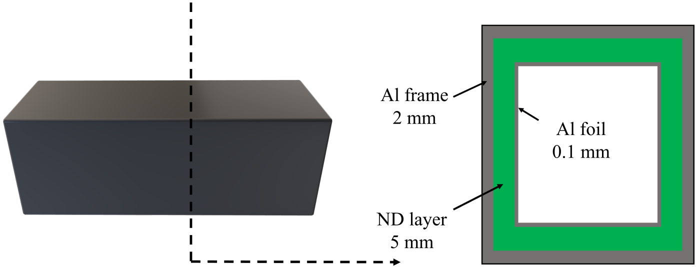
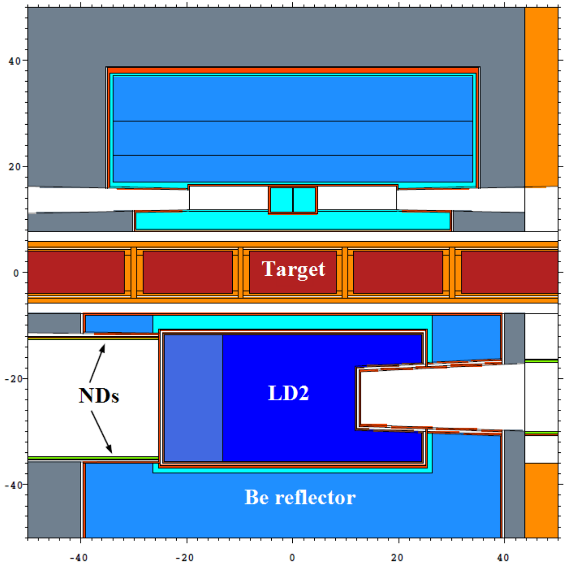
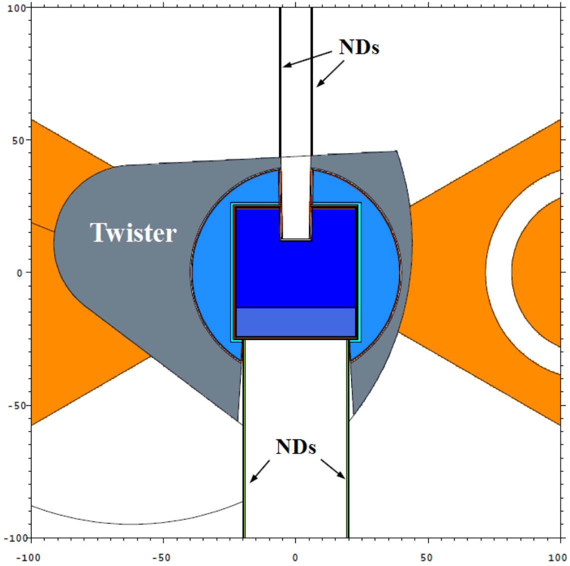
Extraction with NDs

- Guide-like channel $\approx 175\,\mathrm{cm}$ long
- ND layer with thickness of $5\,\mathrm{mm}$ and density $0.6\,\mathrm{g/cm^3}$
- Aluminum frame $2\,\mathrm{mm}$ thick
- Inner aluminum foil of $100\,\mu\mathrm{m}$

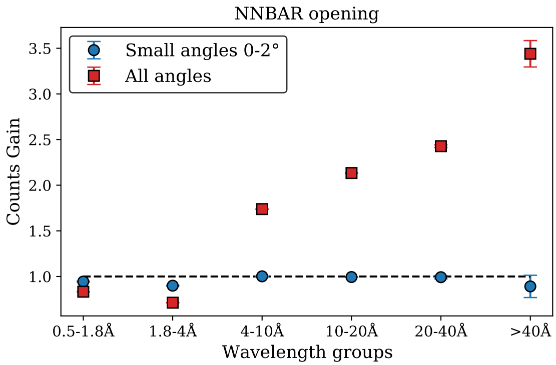


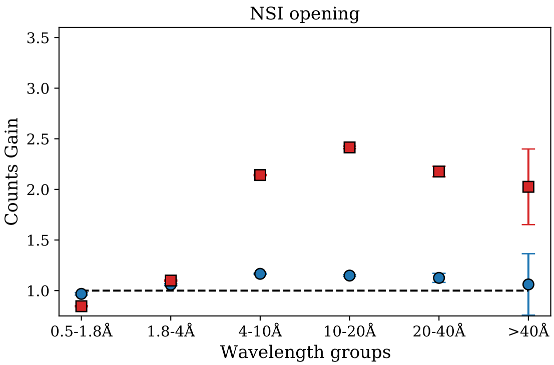
MCNP Model


40
20
0
-20
-40
-40
-20
0
20
40
Target
NDs
LD2
Be reflector

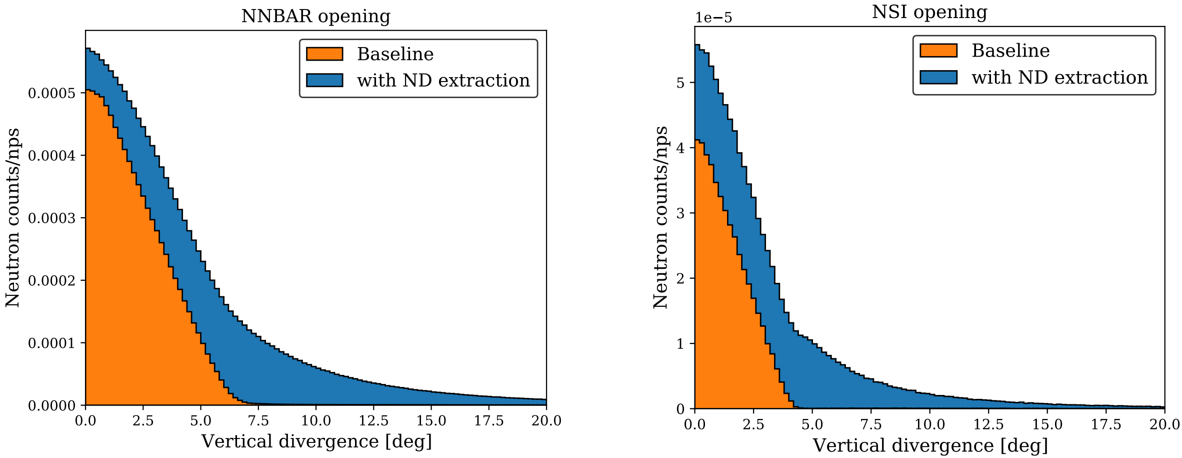

100
50
0
-50
-100
-100
-50
0
50
100
NDs
Twister
NDs

Gains for the two openings





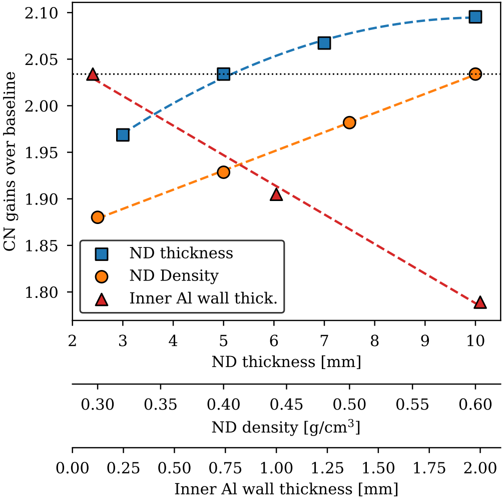
Angular distribution λ > 4Å



Sensitivity of the gains

Key points of the gains sensitivity to changes in the parameters:

- ND layer thickness is the least important parameter
- commercial NDs ($0.3\,\mathrm{g/cm^3}$) is a viable option
- thickness of the inner wall is **critical**. Metal foils as thin as possible.



In-pile solid deuterium source

Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment
Volume 1062, May 2024, 169215

Full Length Article

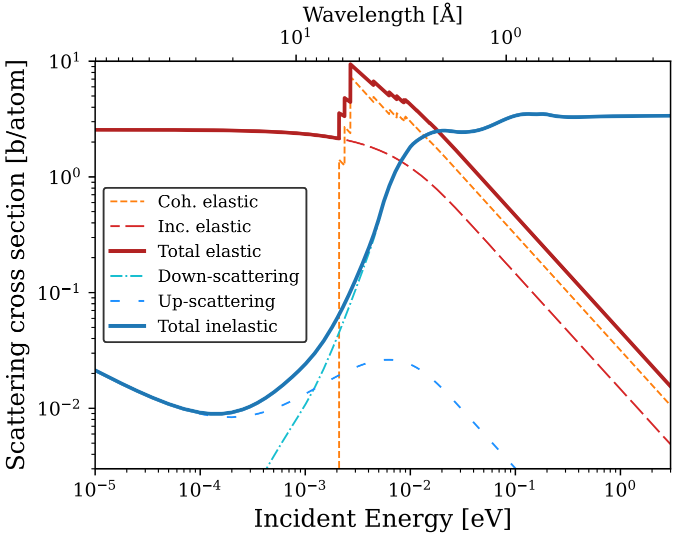
An intense source of very cold neutrons using solid deuterium and nanodiamonds for the European Spallation Source

Nicola Rizzi [a], Ben Folsom [b], Mina Akhyani [c], Mads Bertelsen [d], Peter Böni [e], Yannick Beßler [f], Tomasz Bryś [d], Amalia Chambon [a], Valentin Czamler [g], Bent Lauritzen [a], Jose Ignacio Márquez Damián [d], Valery Nesvizhevsky [g], Blahoslav Rataj [d], Stavros Samothrakitis [h], Valentina Santoro [d], Ha Shuai [i], Markus Strobl [h], Mathias Strothmann [f], Alan Takibayev [d], Richard Wagner [g], Luca Zanini [d], Oliver Zimmer [g]

Solid ortho-deuterium

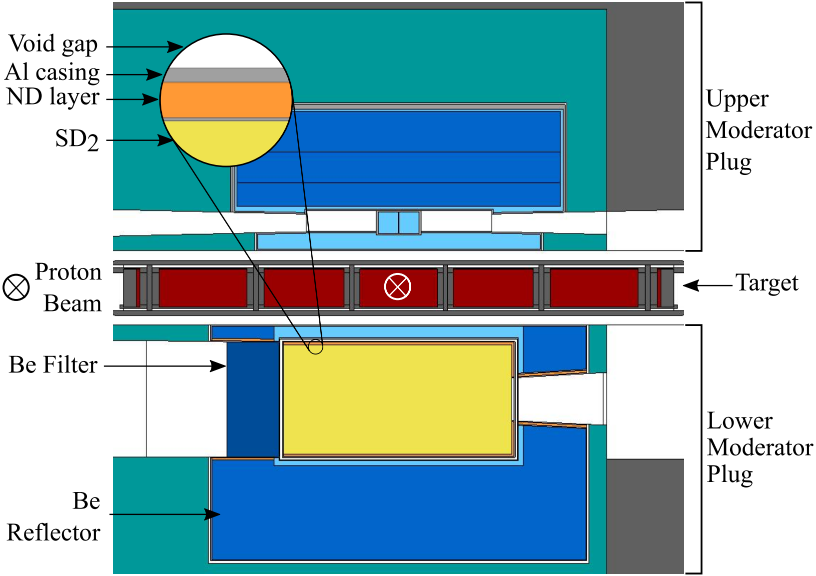
SD_2 at 5 K is an optimal candidate for an in-pile VCN source:

- Small up-scattering cross section for energies less than 20 meV ($\lambda > 2$ Å)
- Large mean free path (≈ 12 cm) at low energies, where the main contribution is incoherent elastic
- Pure o-D_2 (S=0,2;J=0,2,...) has a higher thermal conductivity than n-D_2



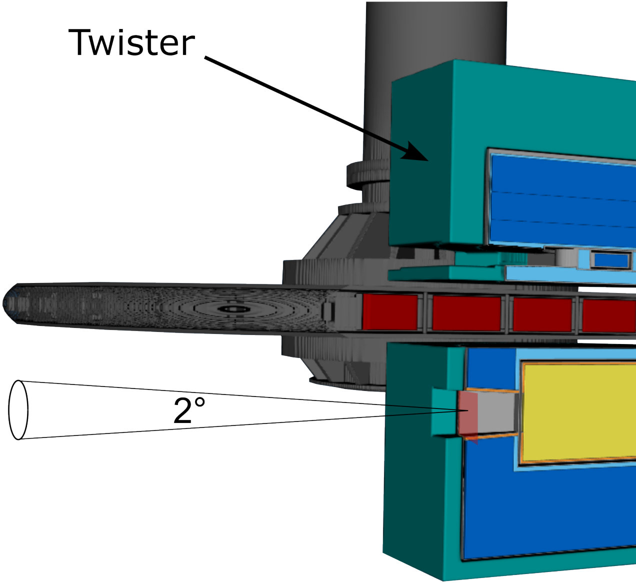
MCNP model

- Same volume as the cold moderator ($\approx 50\,\mathrm{L}$)
- Surrounded by 5 mm of NDs
- external Be filter at 20 K for NNBAR (optional)

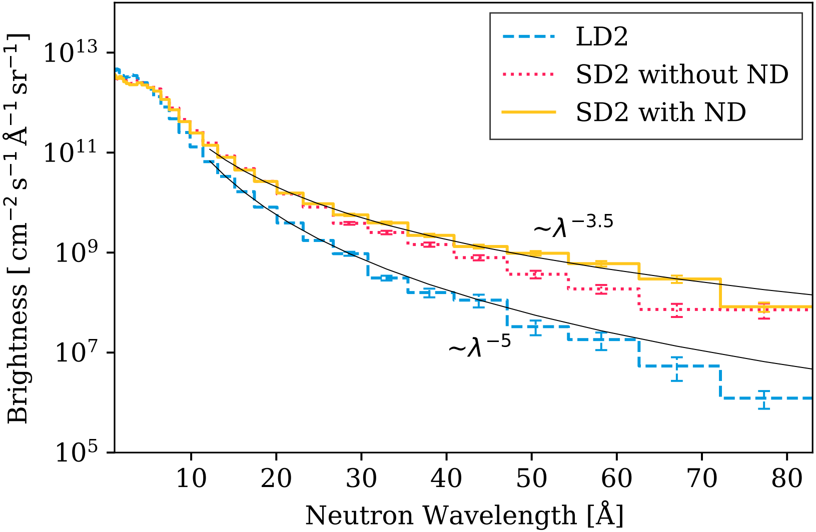


Brightness estimation method

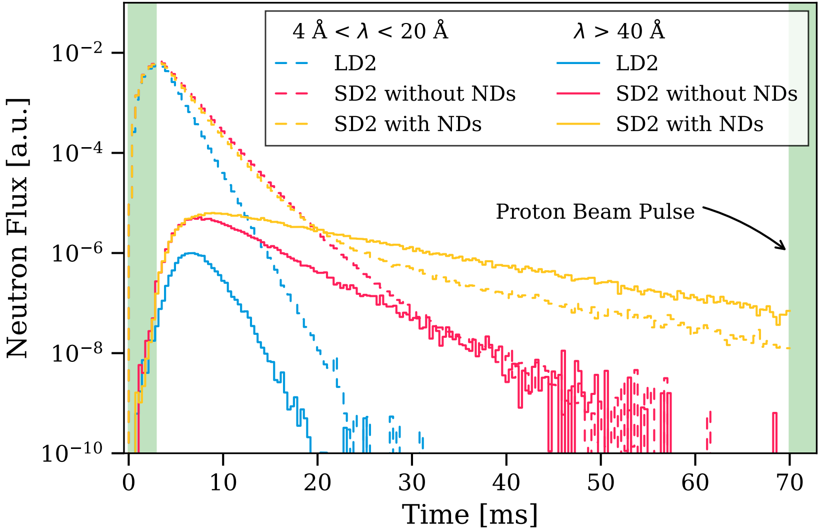
$$B\left[\mathrm{n/s/cm^2/sr/Å}\right] = \frac{C\,[\mathrm{n/proton}]\,P\,[\mathrm{proton/s}]}{S\,[\mathrm{cm^2}]\,\Omega\,[\mathrm{sr}]\,\Delta\lambda\,\left[\mathrm{Å}\right]}$$



Brightness spectra

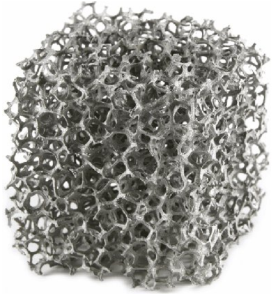


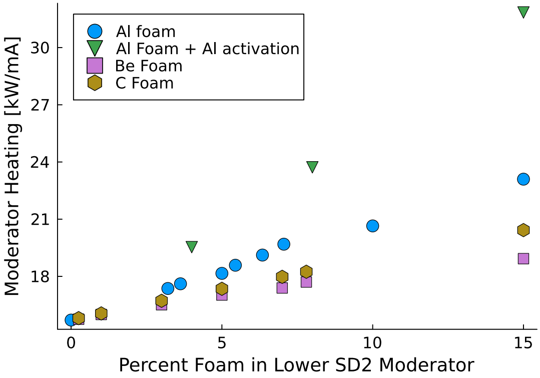
Time response



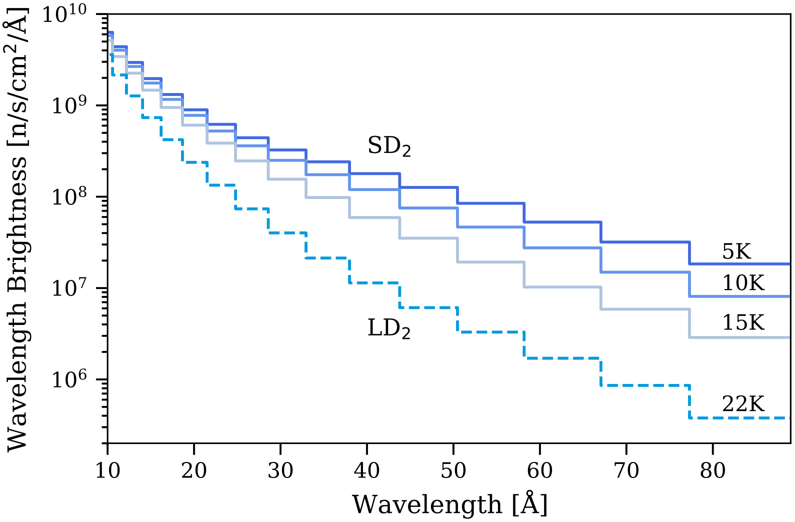
How do we plan to cool it?

- Preliminary calculations by engineering team show it is possible to cool it within the ESS environment at 2 MW beam power by use of metallic foam and liquid-He channel





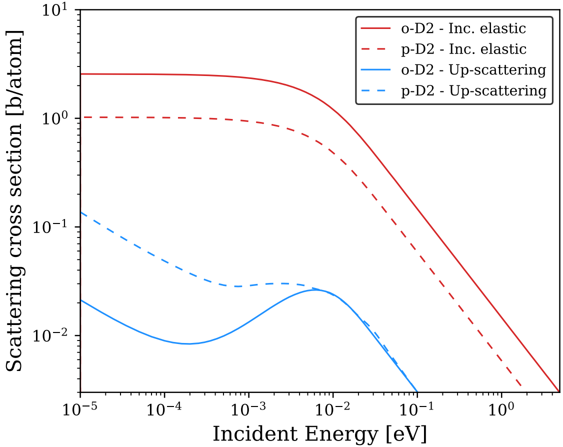
Temperature dependence



Purity

- contamination from HD and para-D_2 does to affect the VCN production
- para-D_2, formed after recombination, increases the thermal resistivity of the crystal Ref.[7]

$$\kappa^{-1} = \frac{c^2}{T^2} \quad (1)$$

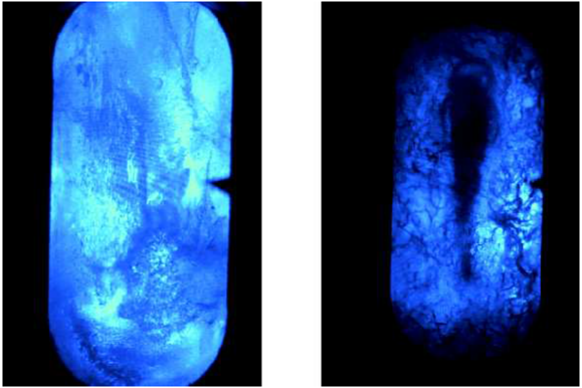


Integrity

- cracks and frost also do not affect VCN production
- they increase the elastic scattering leading to the calculation of sub-optimal dimensions for the moderator

$$P_R = \left| \frac{\sqrt{E_\perp} - \sqrt{E_\perp - \Delta V_F}}{\sqrt{E_\perp} + \sqrt{E_\perp - \Delta V_F}} \right|^2 \quad (2)$$

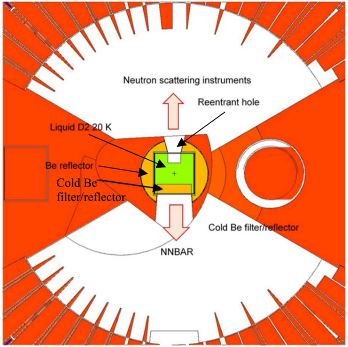
$$P_T = 1 - P_R \quad (3)$$

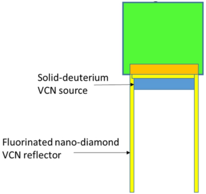


Ref.[2]

Original concept

- Small SD_2 VCN moderator at 5 K
- A ND reflector increases the total flux at the beam port
- thin ND layer between the cold and very cold source
- It is nearly transparent for CNs but traps VCN in the moderating volume



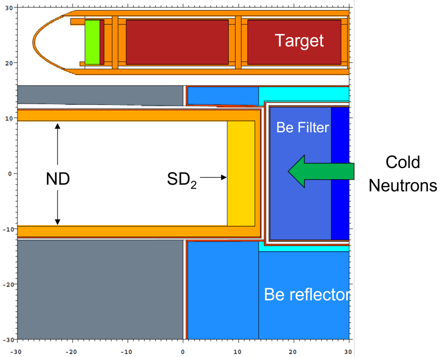


Ref.[5]

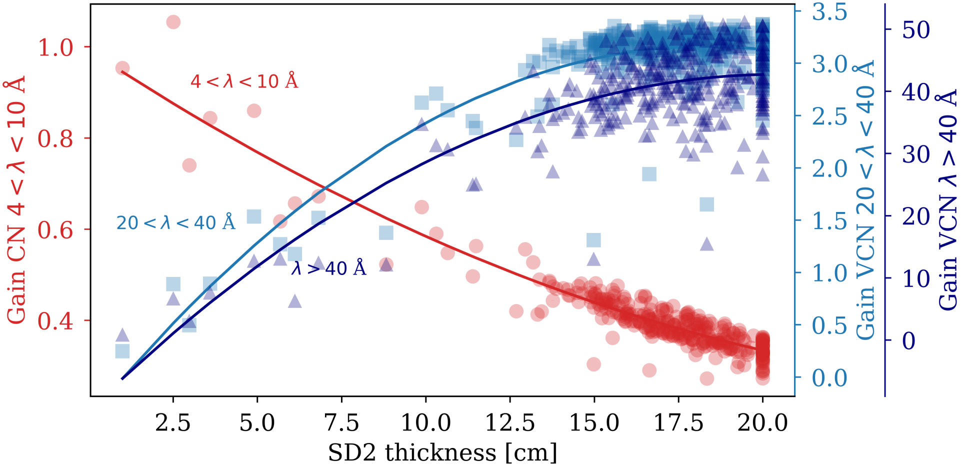
Preliminary Study

Gains	4 Å to 10 Å	20 Å to 40 Å	> 40 Å
Al casing	0.98	0.95	0.86
SD_2 only	0.54	0.71	0.93
NDs only	1.71	0.84	0.44
SD_2 + NDs	0.87	1.76	16.9

Table: Preliminary results at all angles of the SD_2 combined design on the NNBAR side. Gains over LD_2 baseline.



Optimization of the ideal case in Dakota

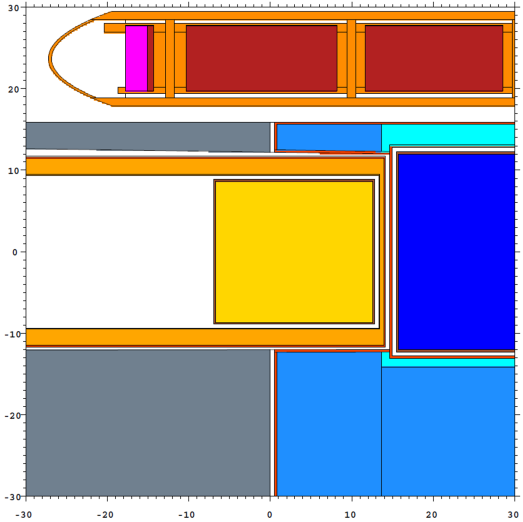


Leaving the ideal case

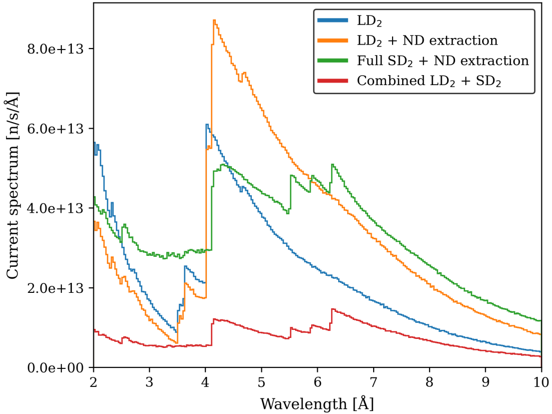
- Converter vessel made of 2 mm Al
- internal reflector walls made of 1 mm Al
- void gaps for the cooling

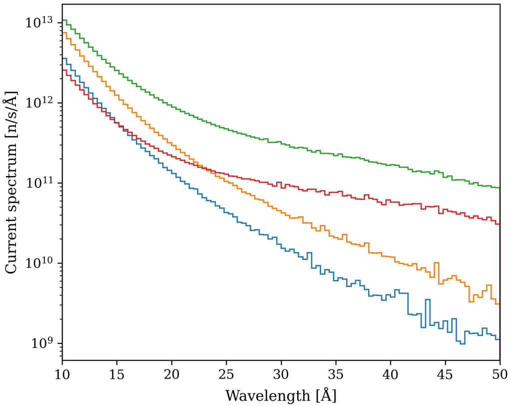
Counts Ratio	4 Å to 10 Å	20 Å to 40 Å	> 40 Å
Over baseline	0.36	1.1	2.6
Over ideal	0.97	0.34	0.05

Table: Preliminary results at all angles of the SD_2 converter design on the NNBAR side. Counts gains over LD_2 baseline.



Designs comparison





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Key takeaways

- SD_2 is a promising material for the first dedicated high-intensity VCN source
- Nanodiamonds, almost transparent in transmission for cold neutron, can be exploited as reflector material for VCNs to increase the performance
- The best option is undoubtedly the full SD_2 moderator. This would replace the established LD_2 moderator as both CN and VCN source
- Cooling appears to be challenging, but:
  1. A VCN source could also operate at higher temperature than 5 K
  2. We should not give up on the possibility to innovate
- Easier solutions are possible, but with trade-offs
- We need tailored instrument concepts to fully take advantage of the characteristic of this first-of-its-kind VCN source

Thank you

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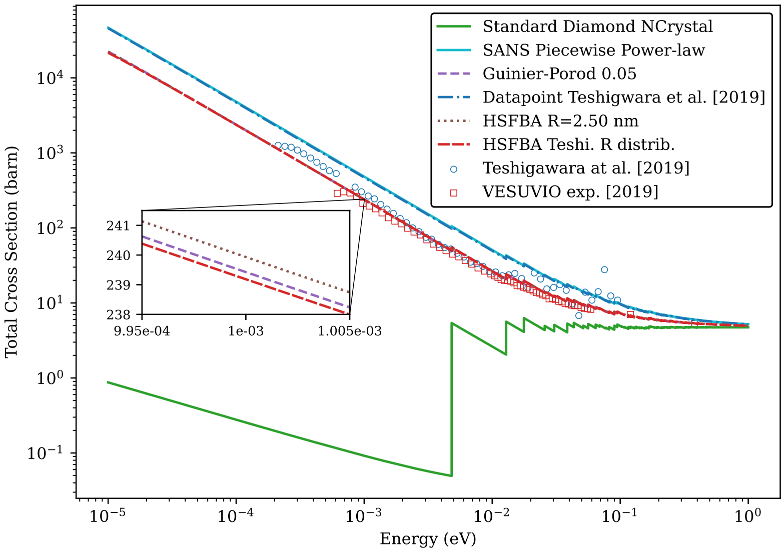
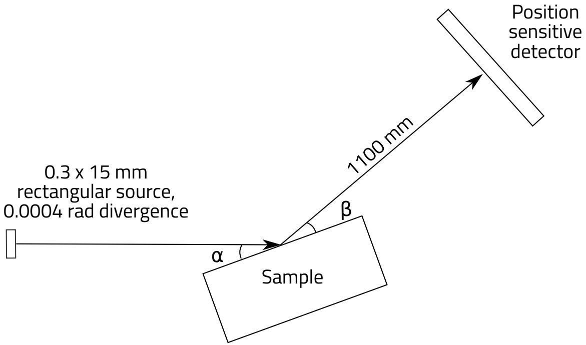
Total Cross Section



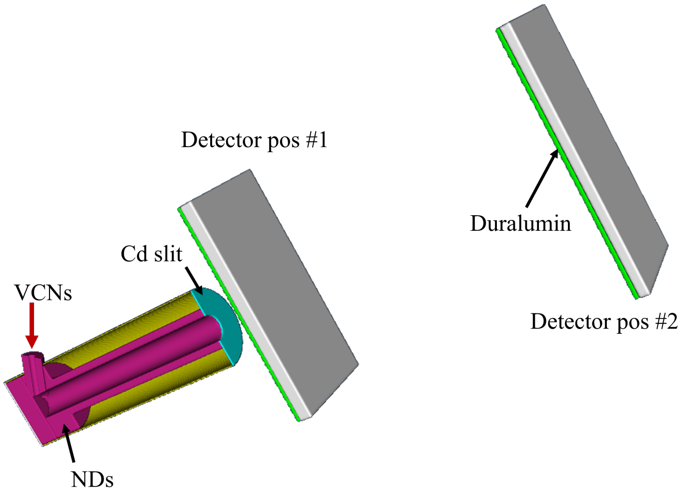
Low-angle reflectometry benchmark

- Comparison with experimental data on quasi-specular reflection of cold neutrons from NDs: Ref.[6]
- Goal: test the plugin in complex configurations where angular distribution of reflected neutrons is essential.



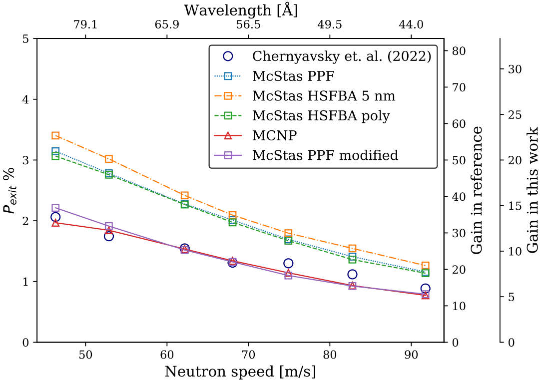
Directional extraction benchmark

- Comparison with experimental data on directional extraction of VCNs from reflector filled with ND powder: Ref.[3]
- Goal: test the plugin in a relevant application for the ESS VCN source.

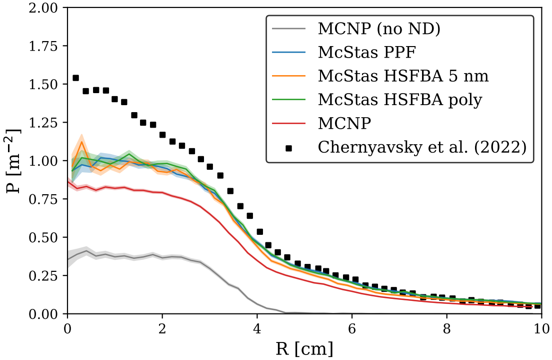


Gain and Specific Probability

Detector close



Detector far



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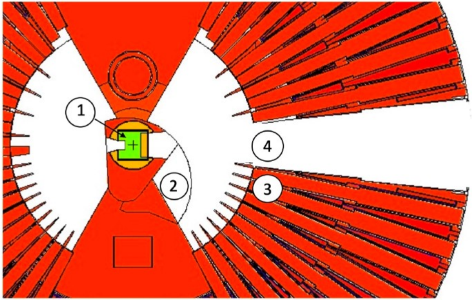
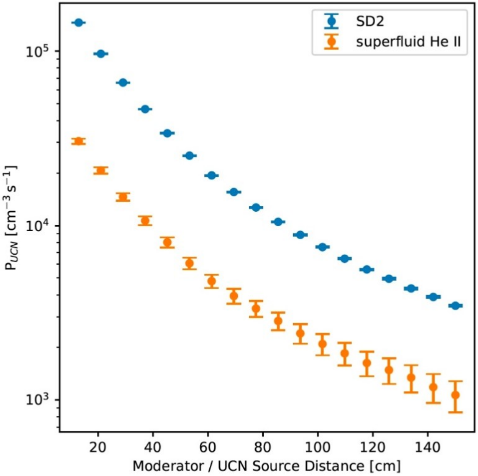
In-pile UCN source

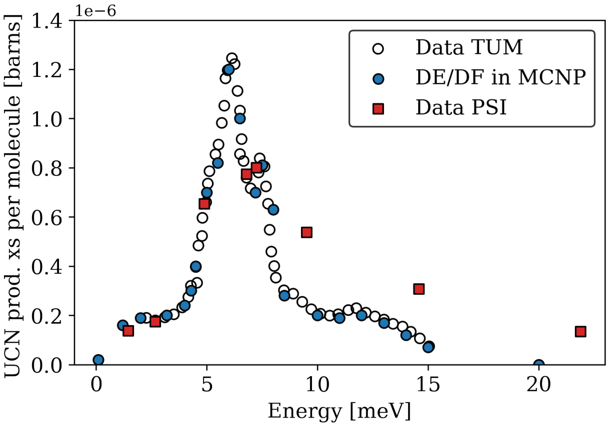
- The rationale behind this concept is to maximize the cold flux delivered to the UCN converter Ref.[10]



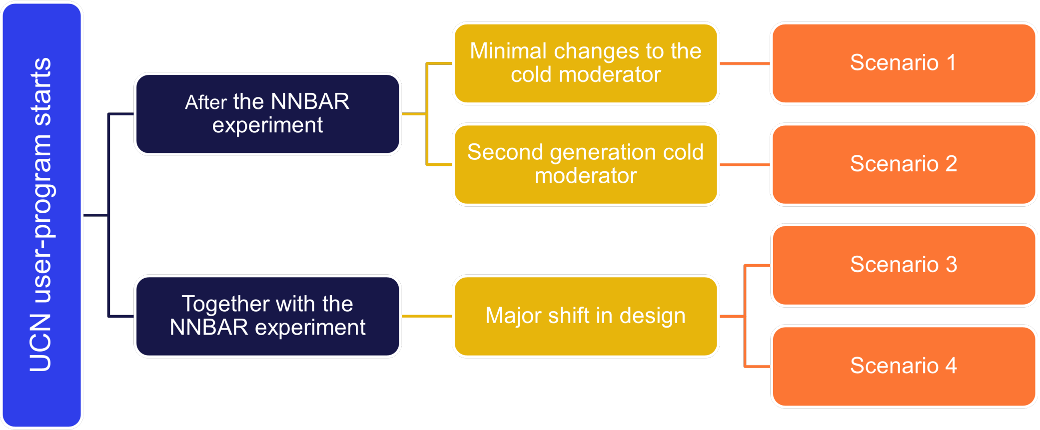


UCN production

- MCNP does not transport UCNs
- Estimation of the cold flux and then multiplied by the UCN production cross section
- The cross section used was calculated in Ref.[4] from a dynamical scattering function extracted from experimental data

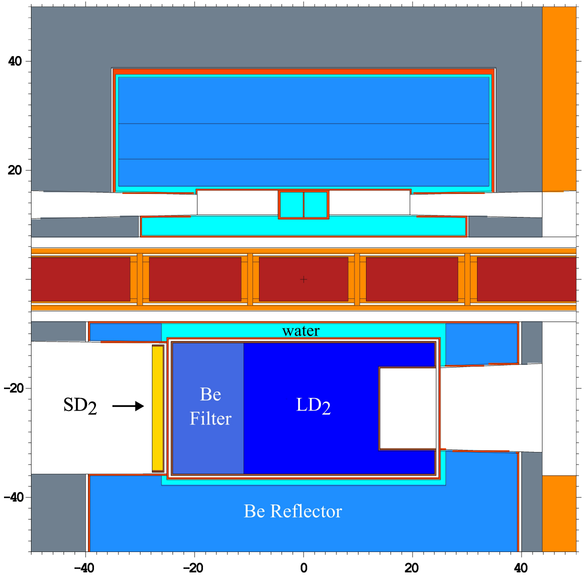


Scenarios UCN user-program



Scenario 1

- The UCN program starts after the NNBAR experiment, and minimal to no changes in the cold moderator are foreseen



Pros

- Independent from the cold source
- Straightforward design

Cons

- Lack of space to accommodate the cooling infrastructure within the pre-designed monolith

Scenario 2

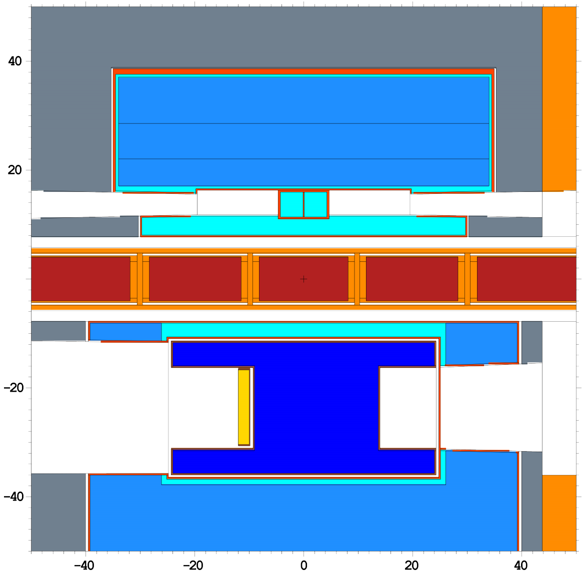
- The UCN program starts after the NNBAR experiment, and a second-generation cold moderator is foreseen

Pros

- Higher UCN density
- Design not too far from the first generation
- Optimization process will now include the FOM for UCN

Cons

- Lack of space is still an issue
- Competing with neutron scattering opening



Scenario 3

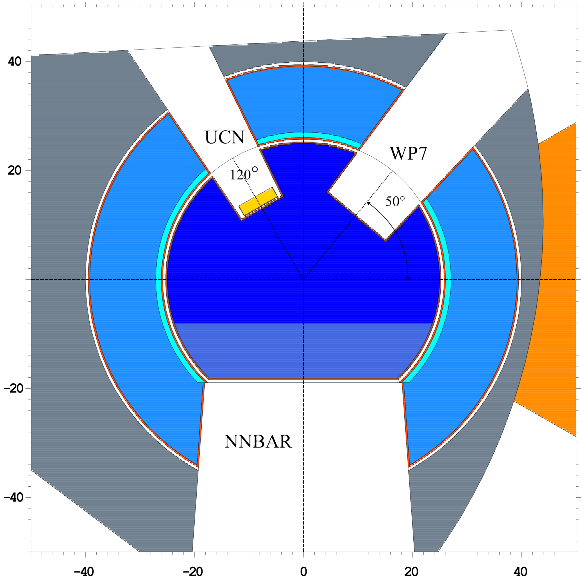
- The UCN program runs together with the NNBAR experiment, which means that a major shift in the design is needed

Pros

- NNBAR, UCN and neutron scattering simultaneously
- Preliminary simulations have shown that the losses for NNBAR and NSI are far from being crippling

Cons

- Many parameters and three figure of merits: complex optimization process



Scenario 4

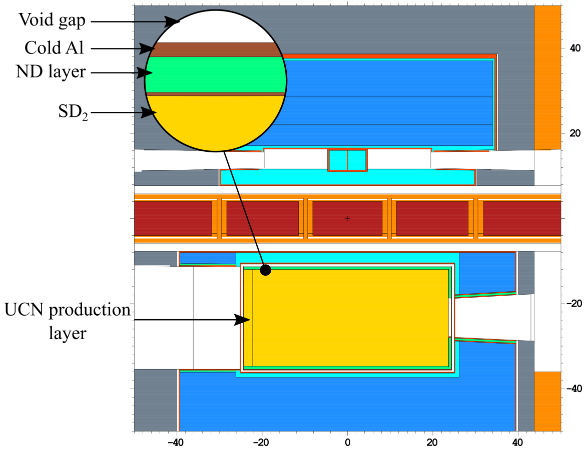
- Large SD_2 source that could serve the NNBAR experiment, but most likely after

Pros

- Cold and very cold source
- UCN produced in the last centimeters

Cons

- New design and engineering challenges



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Efficient Global Optimization

Let F be an expensive black-box function to be minimized (maximized). Let's sample F at the different locations $X = \{x_1, x_2, \ldots, x_n\}$ yielding the responses $Y = \{y_1, y_2, \ldots, y_n\}$.

A Gaussian Process (GP) model (also called Kriging) with a mean function $\mu(x)$ and a variance function $\sigma^2(x)$ is built on the sample and it approximates F.

The Expected Improvement Function (EIF) is used to select the location at which a new training point should be added.

Expected Improvement

The EIF is defined as the expectation that any point in the search space will provide a better solution than the current best solution based on the expected values and variances predicted by the model. To compute it, let us denote:

$$f_{min} = \min\{y_1, y_2, \ldots, y_n\} \tag{4}$$

The EIF can be defined as:

$$E[I(x)] = E\left[\max(y_{min} - \hat{Y}, 0)\right] \tag{5}$$

where $\hat{Y}$ is the GP prediction following the distribution $\mathcal{N}(\mu, \sigma)$. The expected improvement can be expressed in closed form:

$$E[I(x)] = (f_{min} - \mu(x))\Phi\left(\frac{f_{min} - \mu(x)}{\sigma(x)}\right) + \sigma(x)\phi\left(\frac{f_{min} - \mu(x)}{\sigma(x)}\right) \tag{6}$$

where Φ and ϕ are respectively the cumulative and probability density functions of $\mathcal{N}(0, 1)$.

Next point

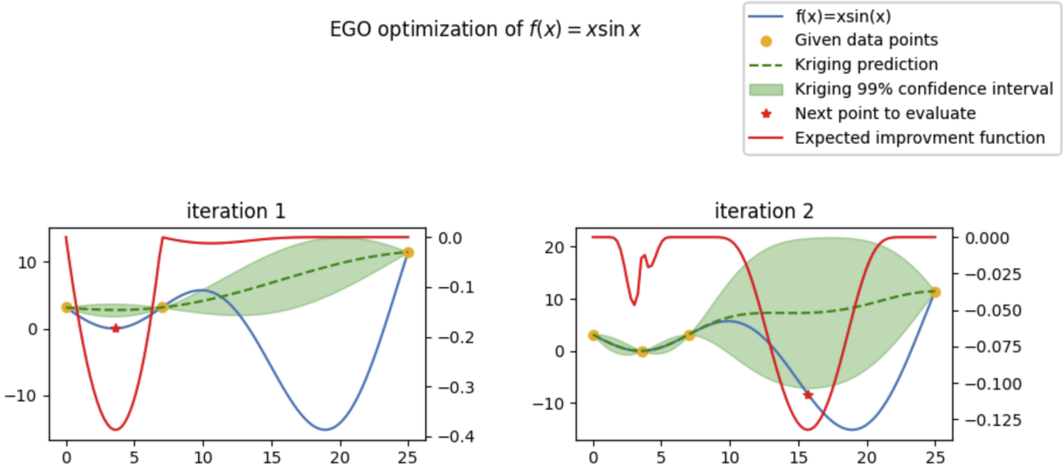
The next sampling point is determined as:

$$x_{n+1} = \arg\max_{x} \left(E\left[I(x)\right]\right) \tag{7}$$

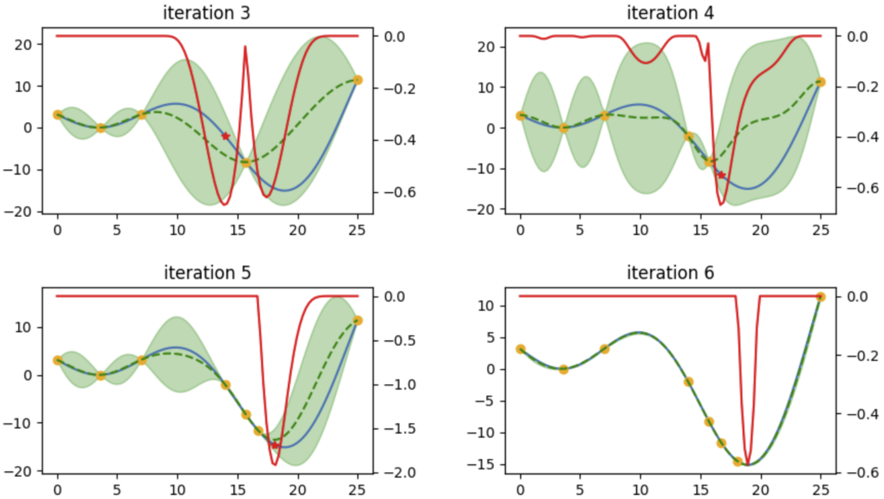
Then the response y_{n+1} of the black-box function F is tested at x_{n+1}, the model rebuilt taking into account the new information gained, and the point of maximum expected improvement searched again.

The point of maximum EIF can also be used as stopping criterion.

Test I


EGO optimization of $f(x) = x\sin x$
f(x)=xsin(x)
Given data points
Kriging prediction
Kriging 99% confidence interval
Next point to evaluate
Expected improvment function
iteration 1
iteration 2

Test II


iteration 3
iteration 4
iteration 5
iteration 6
20
10
0
−10
−20
0.0
−0.2
−0.4
−0.6
0
5
10
15
20
25
−0.5
−1.0
−1.5
−2.0
−5
−15

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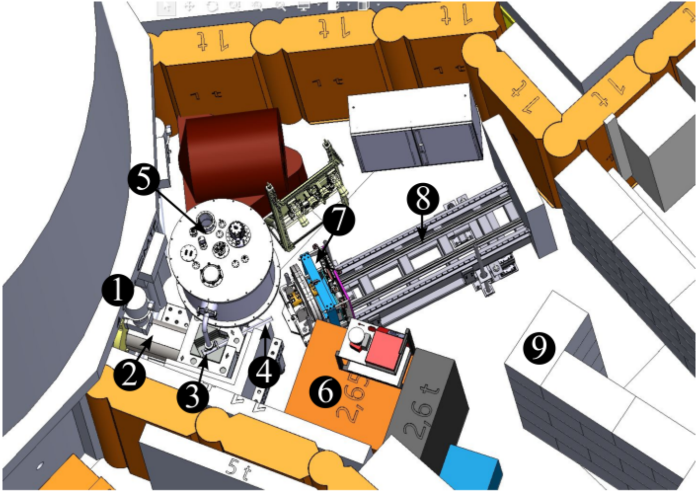
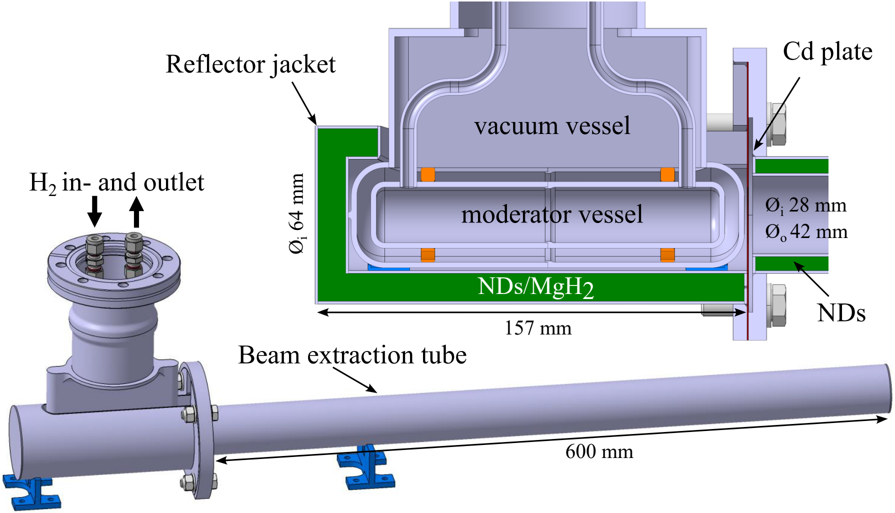
Cold Moderator Test Facility

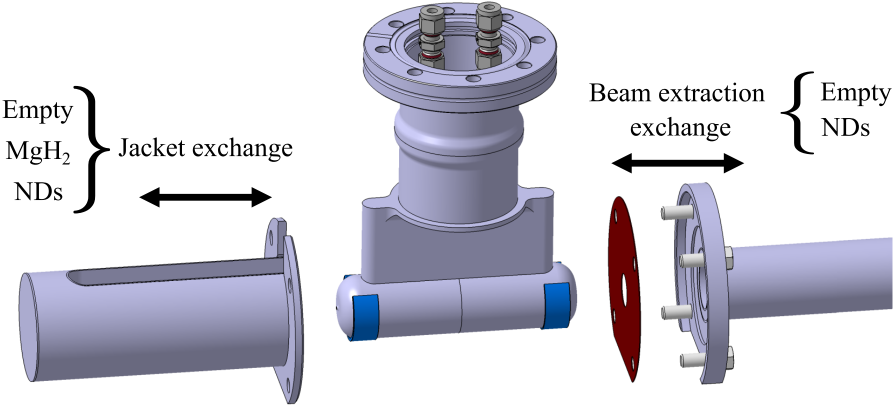


1. Channel shutter
2. Primary carbon steel collimator
3. Lead reflector-moderator block
4. Extraction system
5. Cryo-cooler tank
6. Beam stop
7. Pin-hole assembly
8. Rail support system
9. Bunker's shielding walls

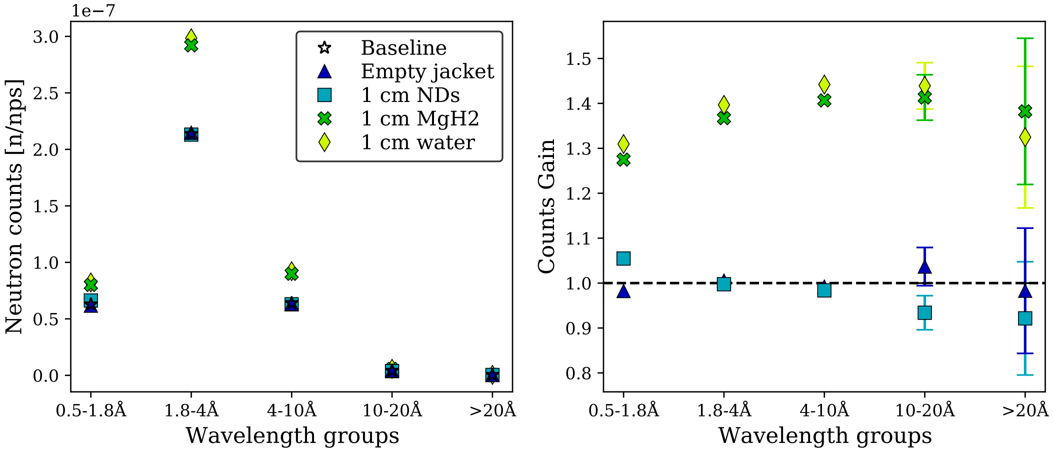
The prototype



Measurements foreseen



Measurements results



Mock-up experiment at BNC

Measurements results