State of the art neutron detection, Helium-3 crisis and potential solutions for neutron scattering applications

K. Zeitelhack

for the

International Detector Initiative

Outline

- Detectors for neutron scattering
- Helium-3 supply shortage
- Initiative to develop alternative techniques to Helium-3 based detectors
    - Scintillation detector technologies
    - Boron-10 converters in gaseous detectors
    - BF_3 – filled detector arrays
- Summary and Outlook

Detectors for neutron scattering applications

What neutrons can do

- No electric charge, magnetic moment μ_n , only weakly interacting with matter
- Neutrons have wave character: λ = 0,03 – 2 nm for E = 900 – 0,2 meV

➔ **Neutrons can probe the atomic structure, magnetic properties and the dynamics of atoms in matter**

Measure (Q,ω) of scattered neutrons

Detector Requirements

- highly efficient
- 2D position resolution: ~1 - 20mm
- time resolution: ~ 1µs
- count rate: 1 – 10^6 Hz
- n / γ-separation
- size: 0,1 – 20 m^2

Typical detectors

- Helium-3 gaseous detectors
- Li-6 based scintillation detectors
- Gd-based Image Plate detectors

Helium-3 use in neutron scattering

Neutron Detection

$n + {}^3He \rightarrow {}^3H + p + 765\ keV$

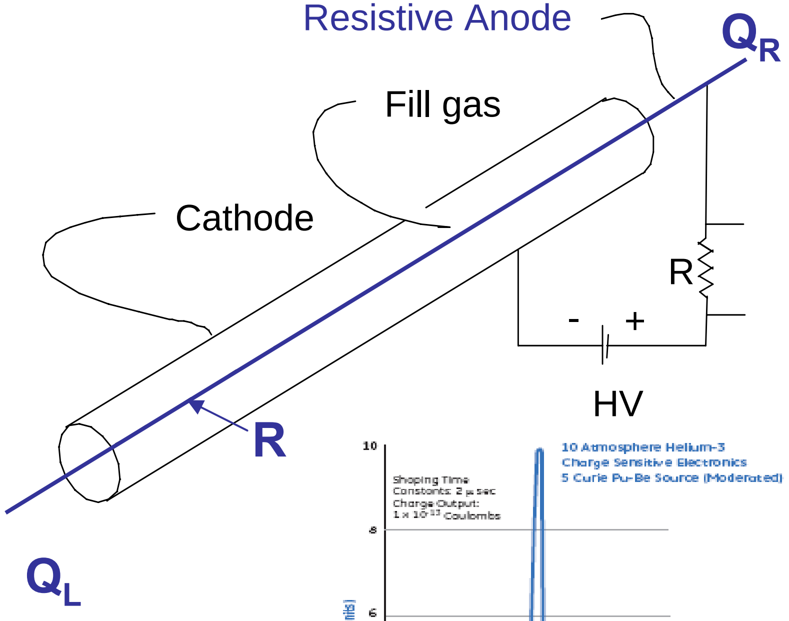
$\sigma = 5330 \cdot (\lambda / 1.8)$ barn

~ 25,000 primary electrons / n

About 75% of detectors for Neutron Scattering use He-3

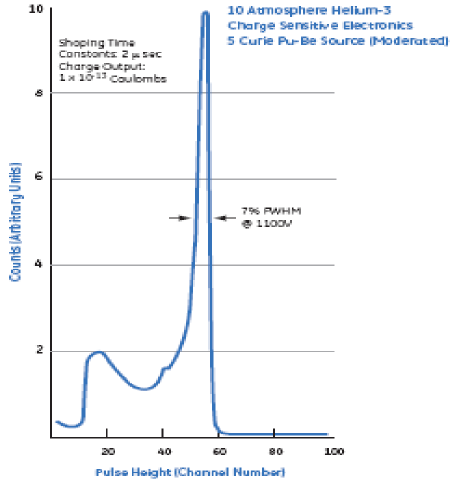
- highly efficient
- good position resolution
- stable
- low background
- very good n / γ-separation
- adequate timing

LPSD



$x \sim (Q_L - Q_R)$

$E \sim (Q_L + Q_R)$



R. Cooper, (ORNL)

Centronic LPSD; 1m, 1”

Some Examples I

Small Angle Scattering

Arrays of LPSDs

- high count rate (~1MHz)
- Δx ~ 5-10 mm
- time resolution Δt ~ µs

SANS1 @ FRM II

128 LPSDs, 1m long, 8mm diameter, 15bar

Diffraction / Reflectometry

MWPC

- 2D position: Δx, Δy ~ 1-2 mm
- moderate count rate (~ 200 kHz)
- time resolution Δt ~ µs

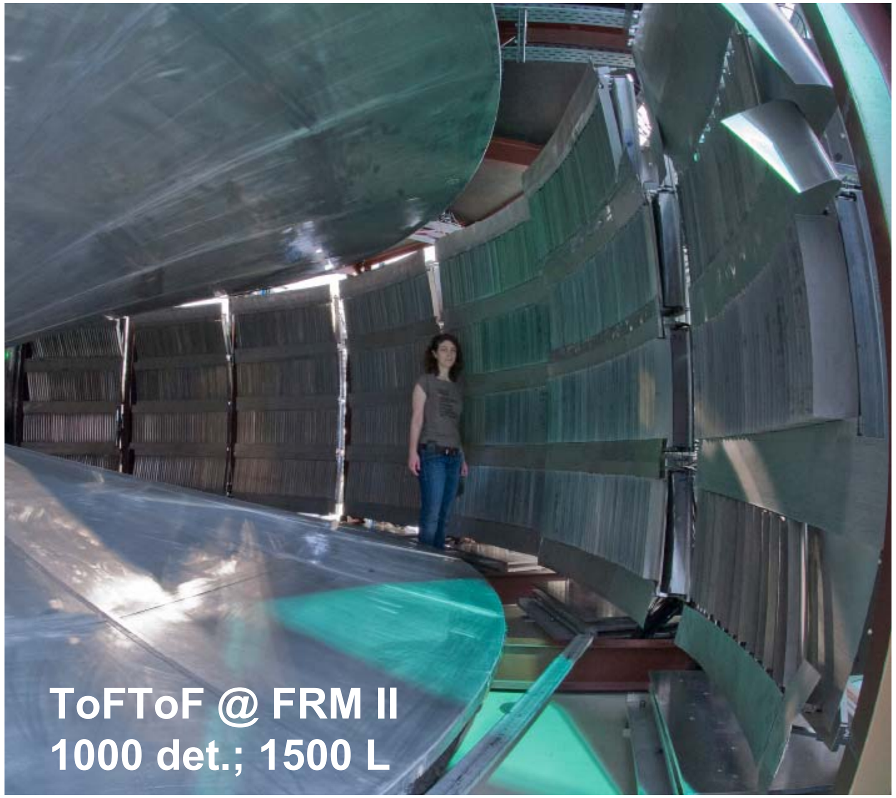
WOMBAT @ Ansto

120°-curved MWPC, 9,5 bar, built by BNL

Some Examples II

ARCS @ SNS
920 LPSDs

IN5 inelastic spectrometer @ ILL
3m long 1” LPSDs, 4,5bar He, 3000 L in total



ToFToF @ FRM II
1000 det.; 1500 L

Typical Inelastic Instruments:

- **Detector area is 15 – 50 m^2**
- **1” LPSDs, 2 – 3m long**
- **He-3 content: 1000 - 4000 L**

Helium-3 supply shortage

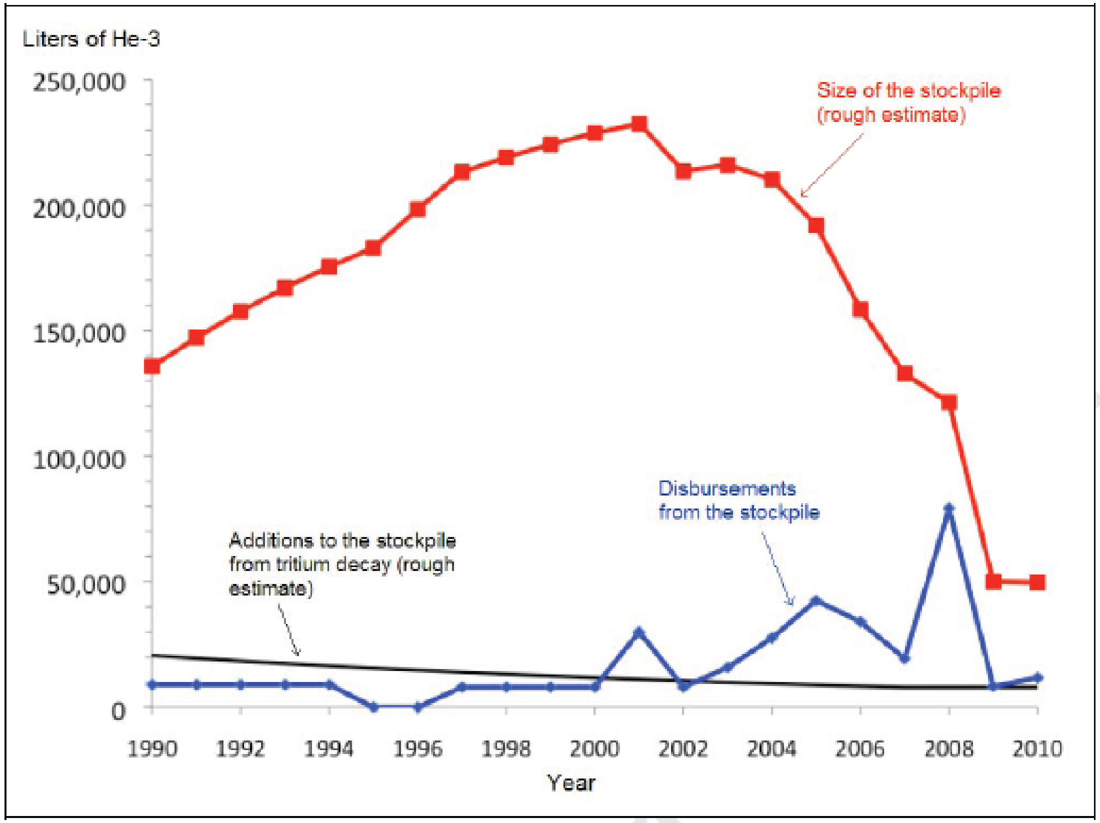
In nature Helium-3 occurs with very low abundance in the atmosphere and in natural gas reservoirs; very expensive to exploit

All available Helium-3 is a by-product of Tritium production for Nuclear Weapons Programs in the USA and Russia !

- Tritium decays via ß-decay into Helium-3 with a 12.3 years half-life
  Helium-3 separated and made available via DOE Isotope Program or Russia
- Tritium production reduced significantly due to disarmament
  US Tritium production stopped in 1988, resumed on small scale in 2003
- Until 2001 He-3 production exceeded demands, Since 2001 increased demand depleted US stock-pile from 235,000L to 40,000 L by 2009 !

Helium-3 supply shortage

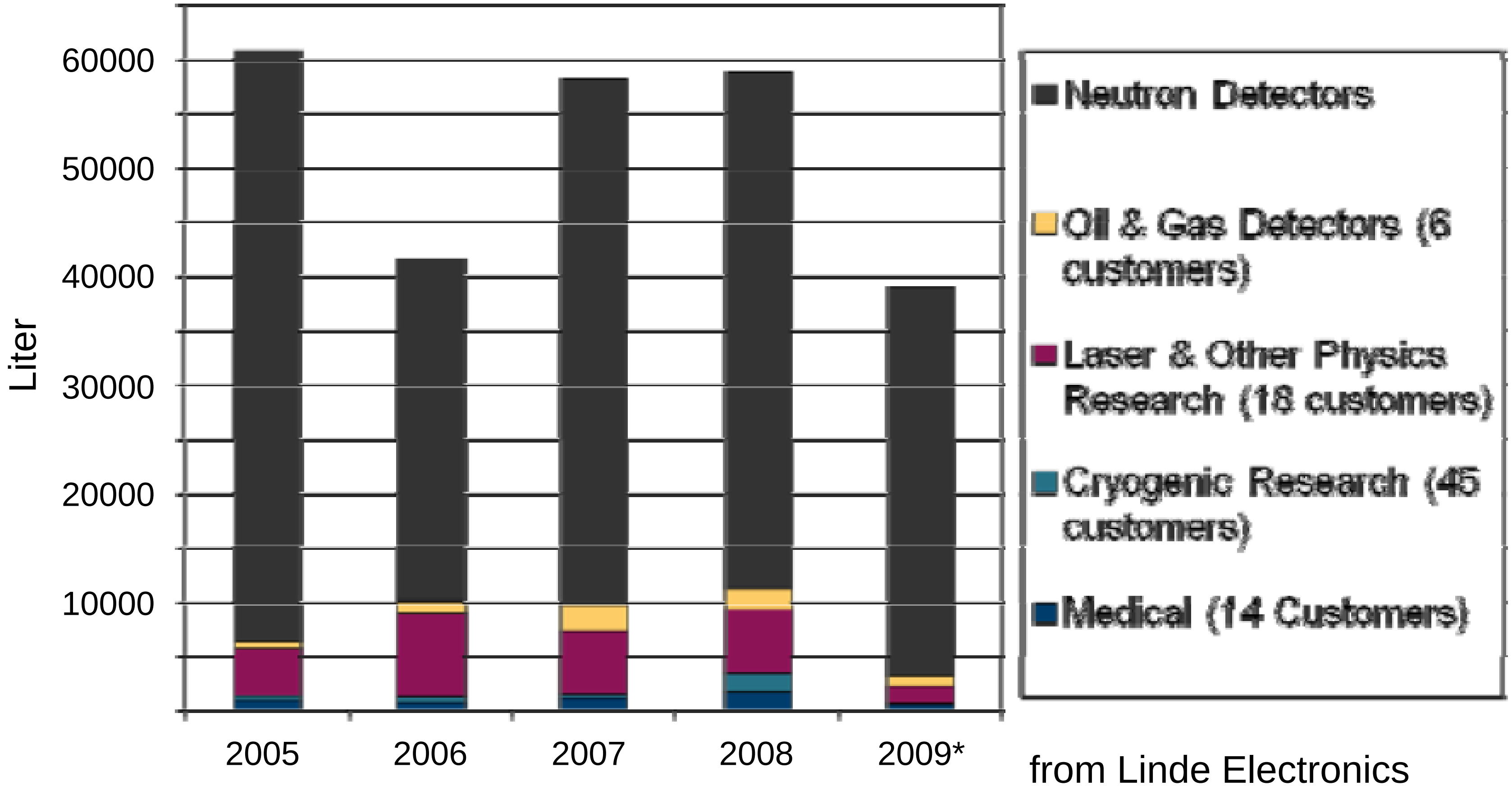
Figure 1. Size of the Helium-3 Stockpile, 1990-2010



Source: Adapted from Steve Fetter, Office of Science and Technology Policy, "Overview of Helium-3 Supply and Demand," presentation at the American Association for the Advancement of Science Workshop on Helium-3, April 6, 2010.

- Security programs and neutron research claim a 5-years demand of ~ 250,000 L !
- DOE stops deployment of Helium-3 in 2009 !

Helium-3 consumption in 2005 - 2009



R. Cooper, (ORNL)

from Linde Electronics
and Specialty Gases

> 80% of Helium-3 used for neutron detectors

Helium-3 Supply - Present Status

Interagency Committee set up to steer rationing and allocation of US Helium-3

- small flow of He-3 resumed from DOE stock-pile
- predicted supply of 8,000 L/Y from the US for the next 6 years;
  presumably similar amount from Russia
- Attempts to exploit additional sources;
  - Retrieve potential 130,000 L He-3 from Tritium extracted from CANDU heavy water reactors and stored in beds
  - Increase dedicated Tritium production by light water reactors

Availability and prices:

- DOE allocated gas for US users only:
  600 $/L for government use / fed. funded research; 1000 $/L for commercial use
- Supply situation from Russia is non-transparent
- Non-US users face uncertain supply and rocketing prices up to 2500 € -3500 € /L

The Detector Initiative

- **The He-3 supply stop prevented the completion and construction of important new instruments at almost all neutron facilities**
- **As a reaction the facility directors decided to initiate a common group of detector experts to prepare a joint R&D program on alternative techniques for large area neutron detectors**
- **Collaboration agreement signed by 9 facilities in 2010**

Consortium Members

ESS	*European Spallation Source*, *Sweden*
FRM II	*Forschungs-Neutronenquelle Heinz Maier-Leibnitz*, *Germany*
HZB	*Helmholtz Zentrum Berlin, Germany*
ILL	*Institut Max von Laue – Paul Langevin, France*
ISIS	*Science and Technology Facilities Council, UK*
JCNS	*Jülich Centre for Neutron Science, Germany*
J-PARC	*Japan Proton Accelerator Research Complex, Japan*
NIST	*Centre for Neutron Research, USA*
ORNL	*Neutron Science Directorate, Oak Ridge National Laboratory, USA*

- **Highest demand to replace large He-3 filled detector arrays**

Detector characteristics to compete

Detector characteristics	10 bar 25 mm diameter ^{3}He
Neutron Efficiency	70% at 1 A
Gamma sensitivity	10^{-6}
Background	10 – 15 counts/ h / m
Width	25 mm
Length	1 - 3 m
Resolution	15 – 25 mm at FWHM
Local rate capability	50 kHz on a pixel
Global rate capability	50 kHz on a tube
Time resolution	1 μs
Area	15 – 40 m^2
Environment	Cryogenic vacuum

Converters for neutron scattering instruments

- Need for a nuclear capture reaction. The kinetic energy range of neutrons in scattering applications is 0.2 meV – 1.5 eV. Too small for proton recoil
- Capture cross section has to steadily cover the broad energy range of neutrons with high efficiency
- Charged particles with sufficient kinetic energy have to be released from the capture reaction to create a detectable electronic signal in the detector
- Signals created by neutrons have to be clearly distinguished from response to gammas and other particles
- Isotope of choice has to be cheap, abundant and easily available
- Converter is stable and usable in “real life” conditions of a detector

Potential Neutron Converters

	Isotope	State	Reaction	Cross Section (b)	Absorb. Length	Product Energies (keV)	Product Range
(✓)	^{3}He	gas	^{3}He(n,p)t	5333	7.59 bar-cm	P:573, t:191	R_p = 0.43 bar-cm CF_4
(✓)	^{6}Li	solid	^{6}Li(n,α)t	940	230μm	T:2727, α:2055	R_t = 130 μm
✓	^{10}B	solid	^{10}B(n,α)^{7}Li	3836	19.9μm	α:1472, ^{7}Li:840	R_α = 3.14 μm
✓	$^{10}BF_3$	gas	^{10}B(n,α)^{7}Li	3836	9.82 bar-cm	α:1472, ^{7}Li:840	R_α = 0.42 bar-cm
✗	^{nat}Gd	solid	^{nat}Gd (n,γ)	49122	6.72μm	Ce:29-182 (86.5%)	Λ_{ce} =12.3 μm

for 25meV Neutrons

Data from Th. Wilpert, (HZB)

Development lines of Detector Initiative

Scintillation Working Group (*ISIS*, *JCNS, J-Parc, NIST, ORNL*)

Investigation and development of scintillation detector technologies for large area detectors

- Build on experience with detectors based on $ZnS:^{6}LiF(Ag)$ or $ZnS:^{10}B_2O_3(Ag)$ scintillators read out by coded arrays of clear or wavelength shifting fibres
- Investigate scintillators, optics, light readout devices, encoding schemes

^{10}B-Working Group (*ILL*, *ESS, FRM II, HZB, ORNL*)

Development of solid ^{10}Boron multilayer arrangements in gaseous large area neutron detectors

- Study ^{10}B-coating processes
- Investigate and optimize design and fabrication of a multilayer detector in view of performance and cost

BF_3-Working Group (*HZB*, *FRM II, ILL*)

Investigate BF_3 as a potential fast and easy replacement of ^{3}He

- Study gas properties, performance and limitations of BF_3
- Investigate safety issues for large scale use

^{10}BF$_3$ Proportional Counters

Simple Solution ! Just replace ^{3}He by ^{10}BF$_3$ in present detector designs
Works as well in a proportional counter, LPSD, MWPC

- Energy deposit 2.3 MeV / neutron

  → *large detector signals; good position resolution ($\Delta L/L = 0.6\%$)*

  → *excellent n / γ-separation ($< 10^{-6}$)*

- Cross section = 72% of Helium

BUT !

- In the past efficiency was limited by low pressure (< 1.3 bar) operation

  → *several detector rows needed to achieve adequate efficiency depending on neutron wavelength*

- Corrosive and highly toxic

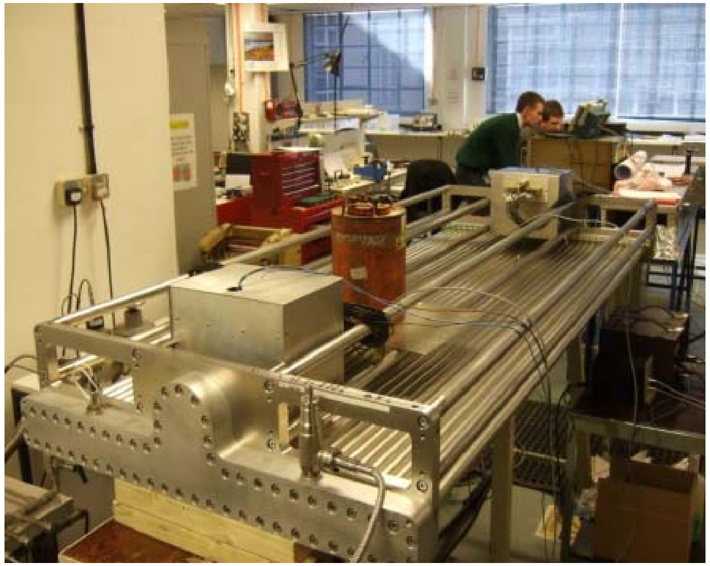
- High voltage increases rapidly with pressure

→ **High pressure operation feasible ?**
Solution for cold neutrons only ?

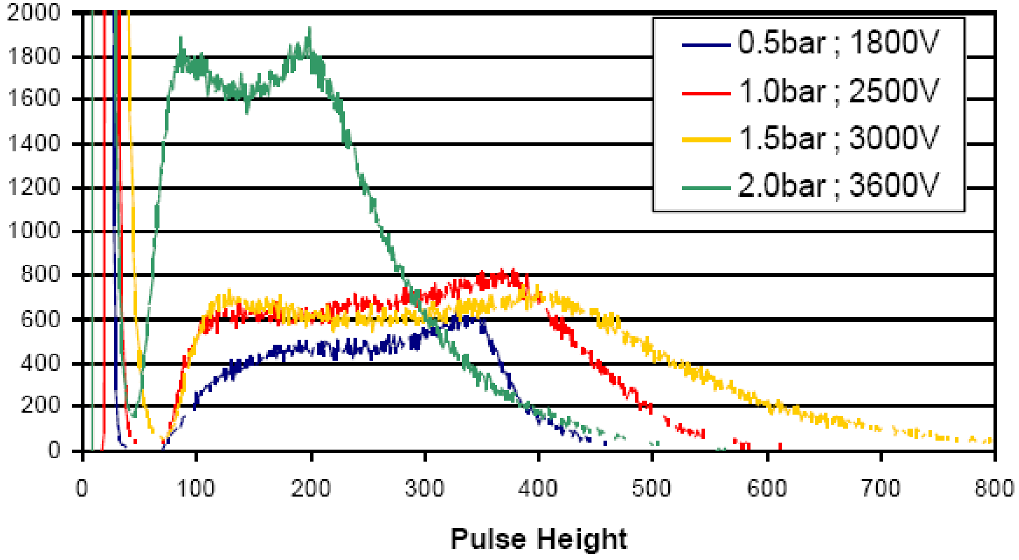
LPSD filled with $^{10}BF_3$ at high pressure

Test of ILL- IN5 prototype module filled with BF_3

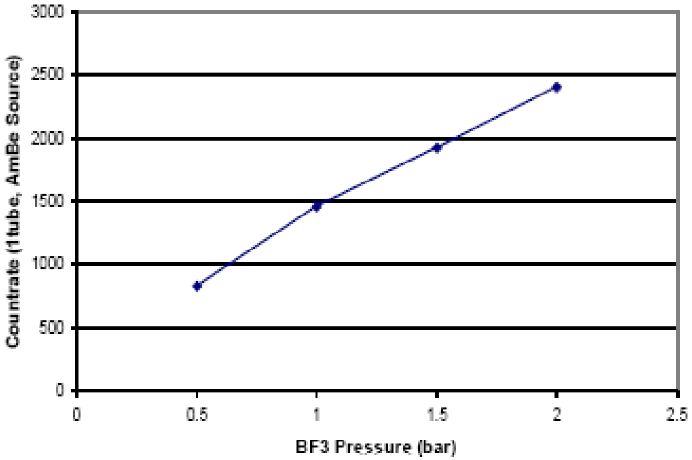
- 32 tubes; 1” x 2m
- BF_3 pressure up to 2bar



Spectra @ PSD Gain for various BF3 Pressures

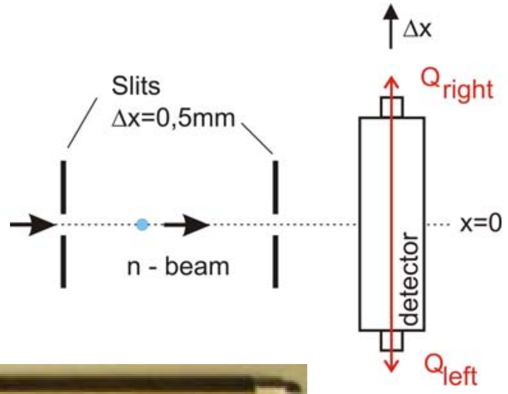


Count rate vs BF3 Pressure



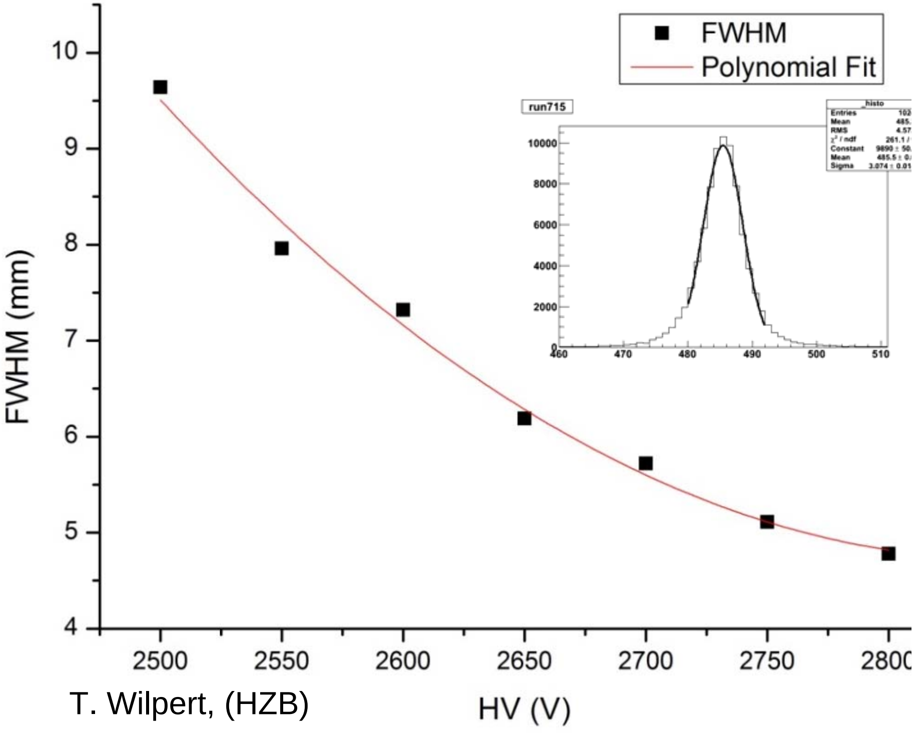
B. Guerard, M. Platz, (ILL)

LPSD filled with $^{10}BF_3$ at high pressure

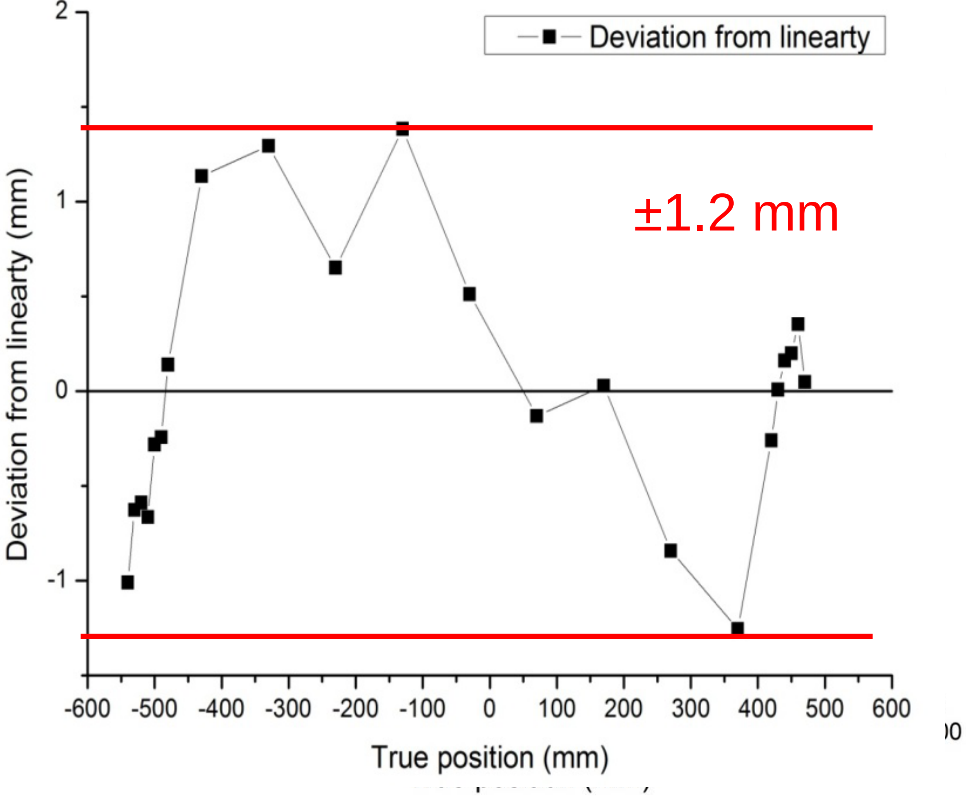


Evaluation of PSD at FRM II and HZB

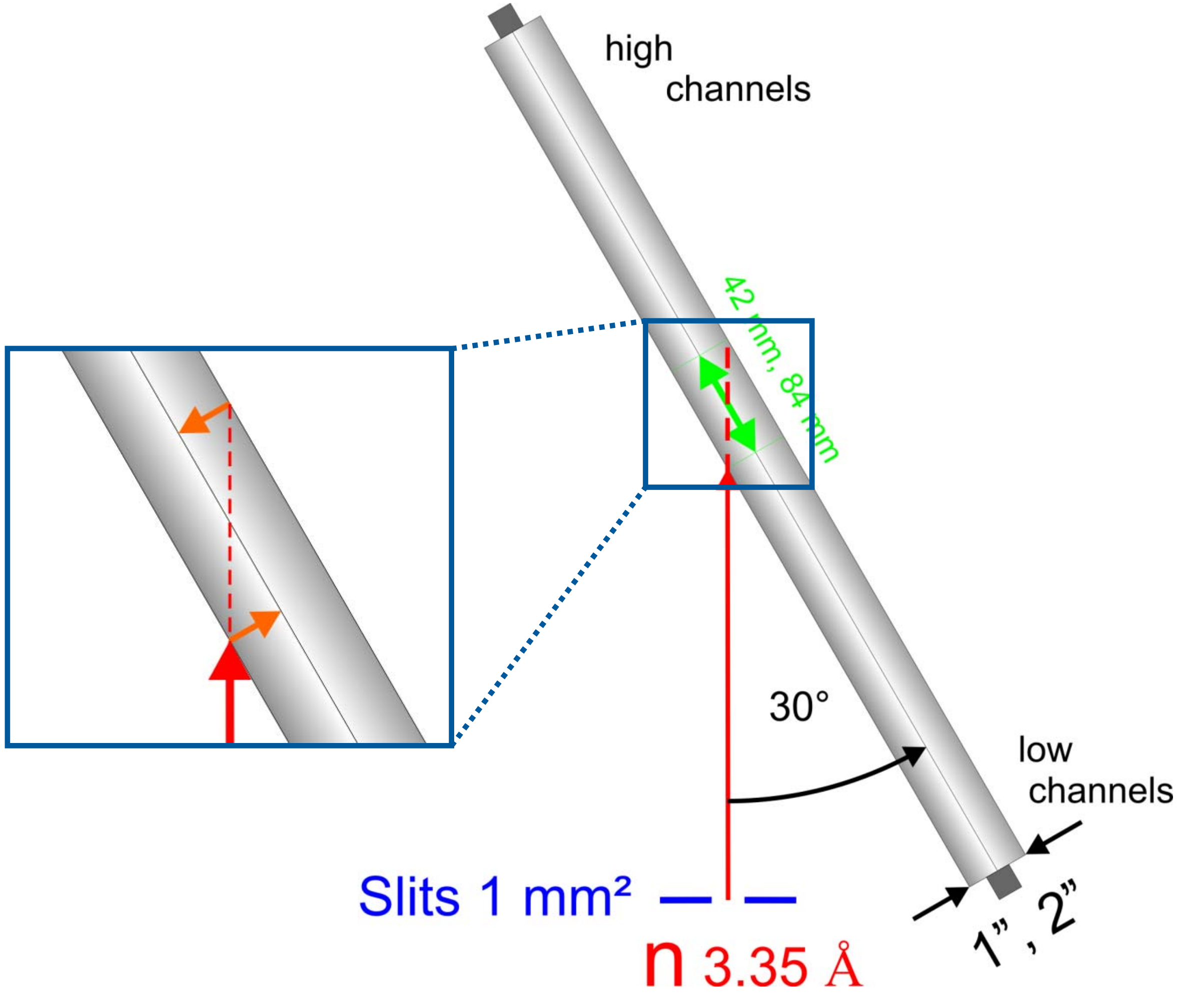
- 1" x 1m LPSD (made by Centronic, UK)
- 1.87 bar, 2800 V max. tested



T. Wilpert, (HZB)

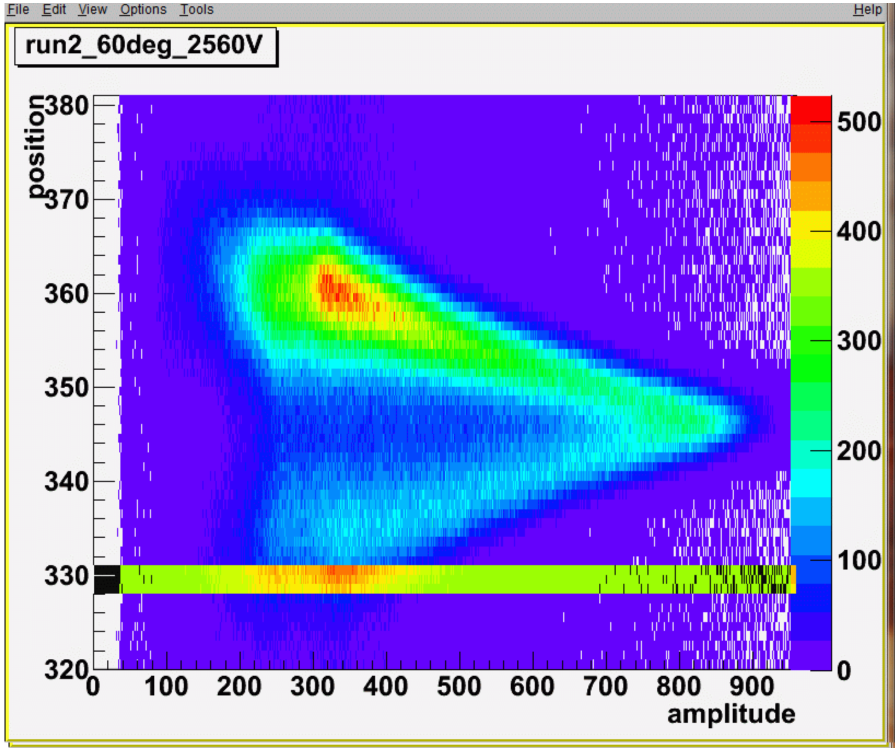


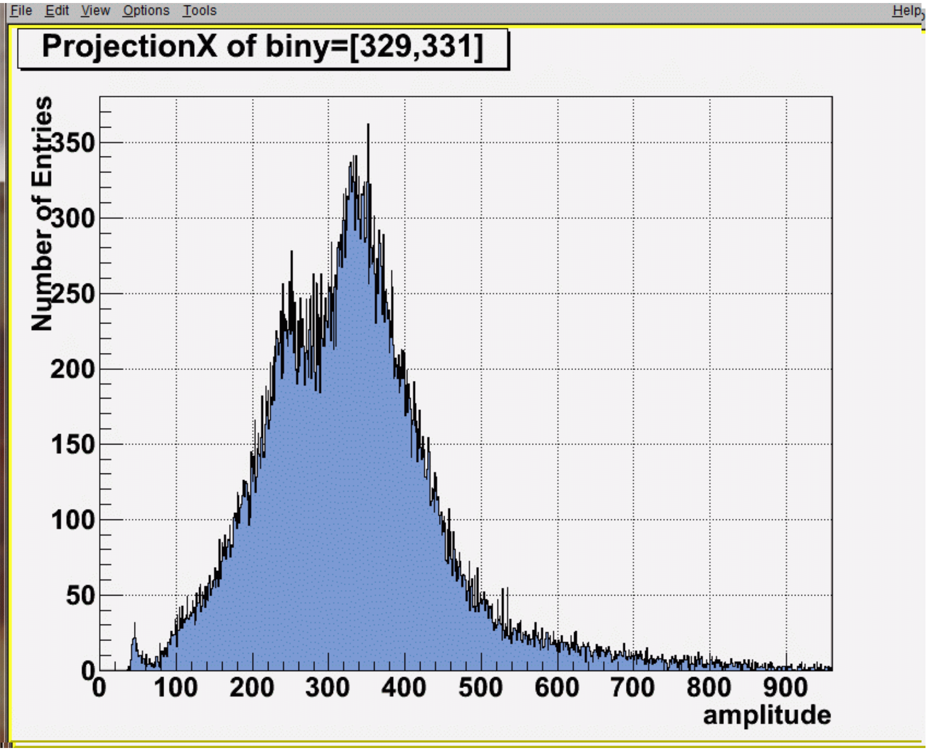
Pulse Height vs Amplitude @60° Inclination



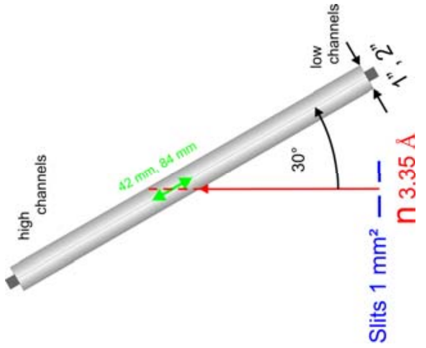
1 mm² spot in tube center (low intensity)

BF_3 Tube (1“) – 60° Run   Pulse Height Distribution

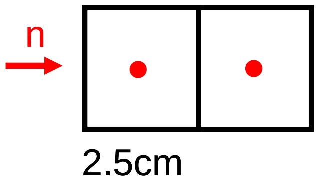


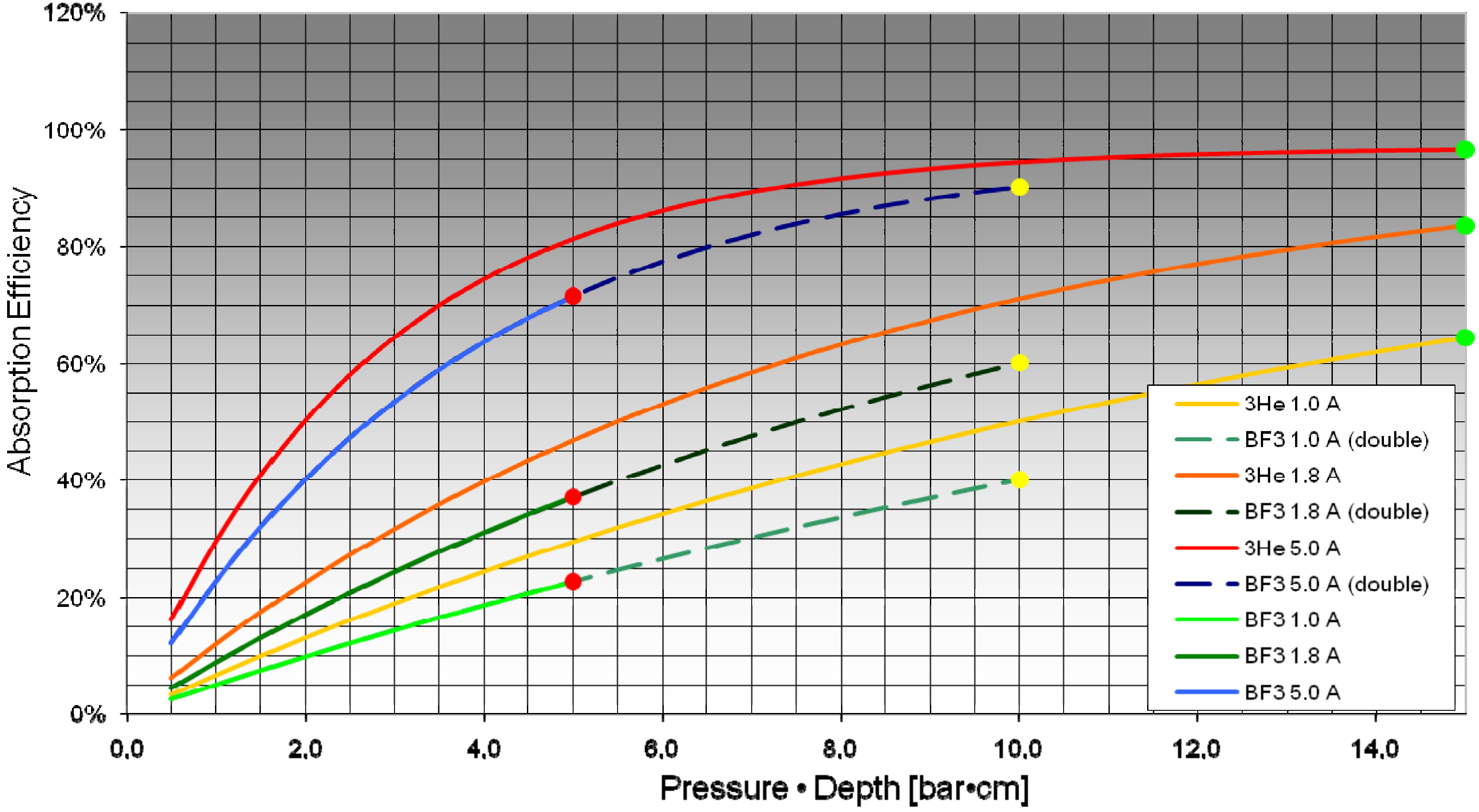


HV 2560 V, Δx = 8 mm fwhm



Neutron absorption efficiency of $^{10}BF_3$





T. Wilpert, (HZB)

NEAT – ToF spectrometer at HZB

40 m² active detector area

544 LPSDs (17 Modules)

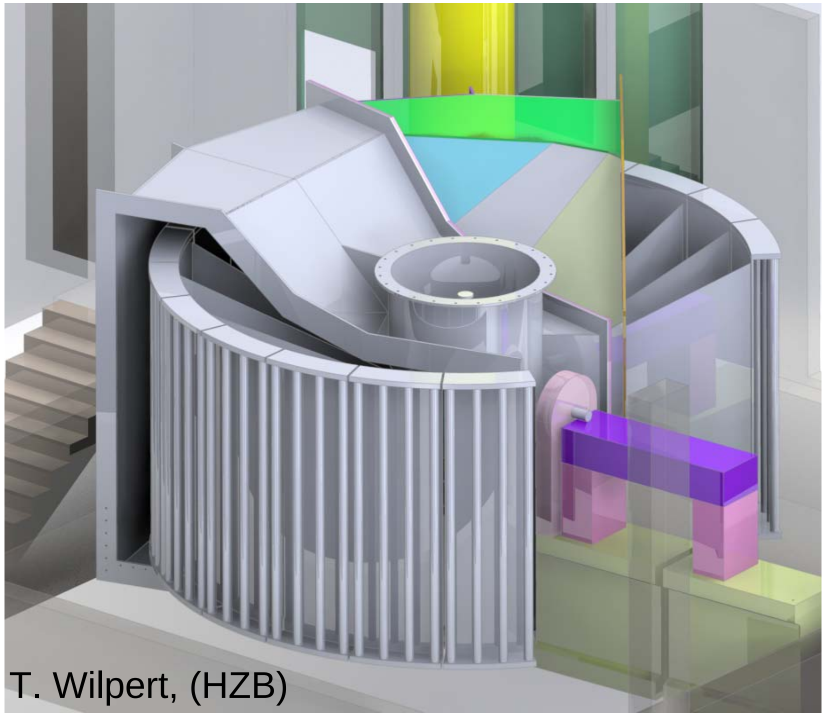
Pixel resolution 2.5 x 2.5 cm^2

ToF resolution < 30 µs @ 5Å

1” width, 3 m long, 90%@ 5Å

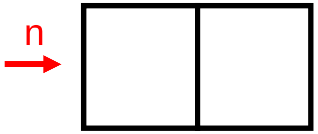
Radius of detection plane 3 m

In-plane angle 280°



T. Wilpert, (HZB)

2 x 2.5 x 2.0 cm²



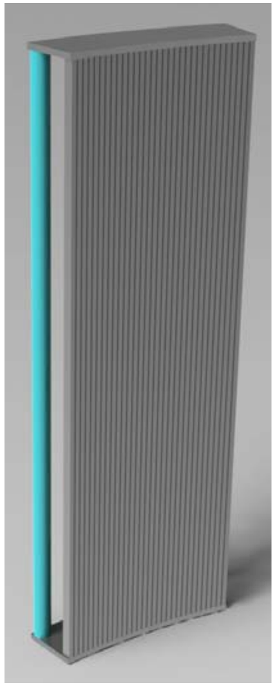
BF_3 pressure **2.5** bar

Efficiency@5.00 Å 90%

Detector module with 32 double-tubes

Backside

Front-side



^{10}B-converter in gaseous detectors

Neutron Detection

$$n + {}^{10}B \rightarrow {}^{7}Li\ (0.84MeV) + \alpha\ (1.47MeV) + \gamma\ (0.48MeV) \quad (93\%)$$

$$\rightarrow {}^{7}Li\ (1.02MeV) + a\ (1.78MeV) \quad (\ 7\%)$$

Why using Boron as solid converter ?

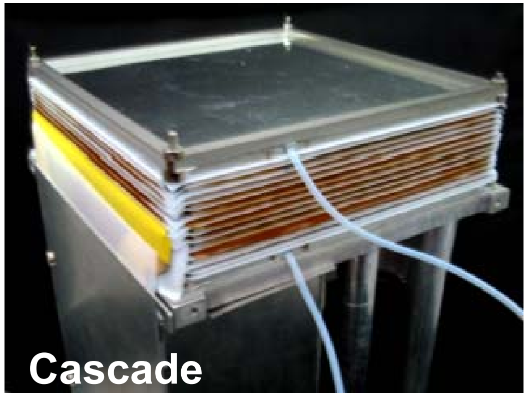
- B_4C stable, not hygroscopic (e.g. as Li)
- large charge signal in detector
- 96% enriched ^{10}B available

State of the art detectors

- B-coated straw tubes

  4mm straws by *"Proportional Technologies"*
- stack of B-coated GEM foils

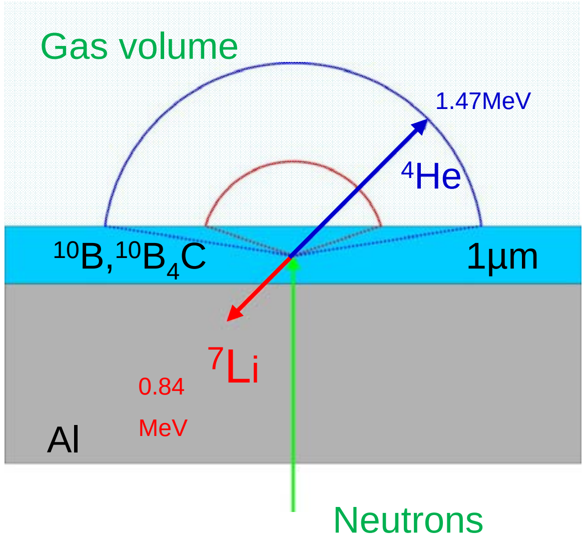
  20x20cm^2 GEM by *"CASCADE", Univ. Heidelberg*



Many open questions

Not adapted for large area, efficiency, homogeneity, robustness, cost

^{10}B-converter in gaseous detectors



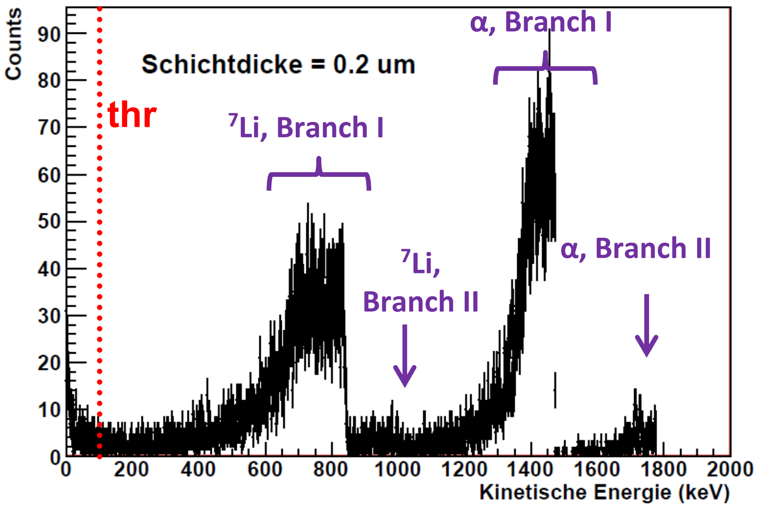
Boron:

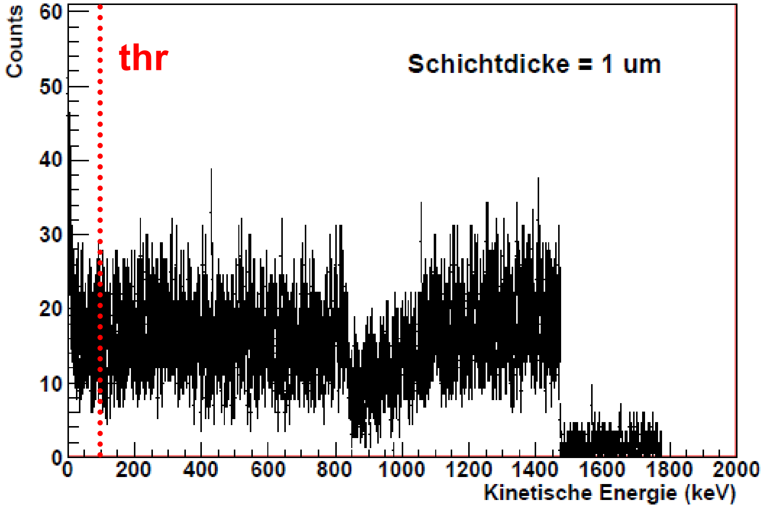
λ_{abs} for therm. neutrons: 20 µm

Range: α = 3.14 µm; Li = 1,53 µm

Pulse height spectra simulated with GEANT4

I. Stefanescu, (FRM II)



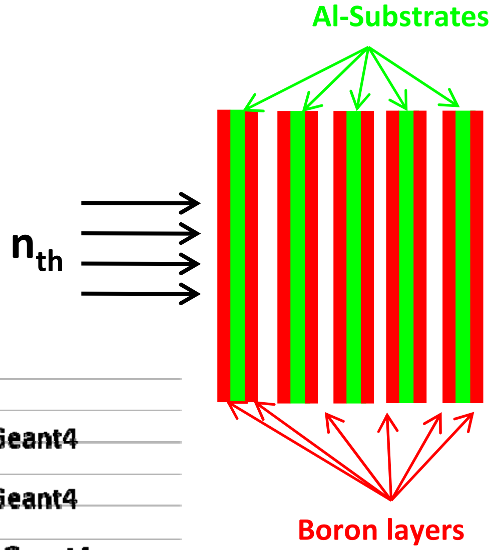


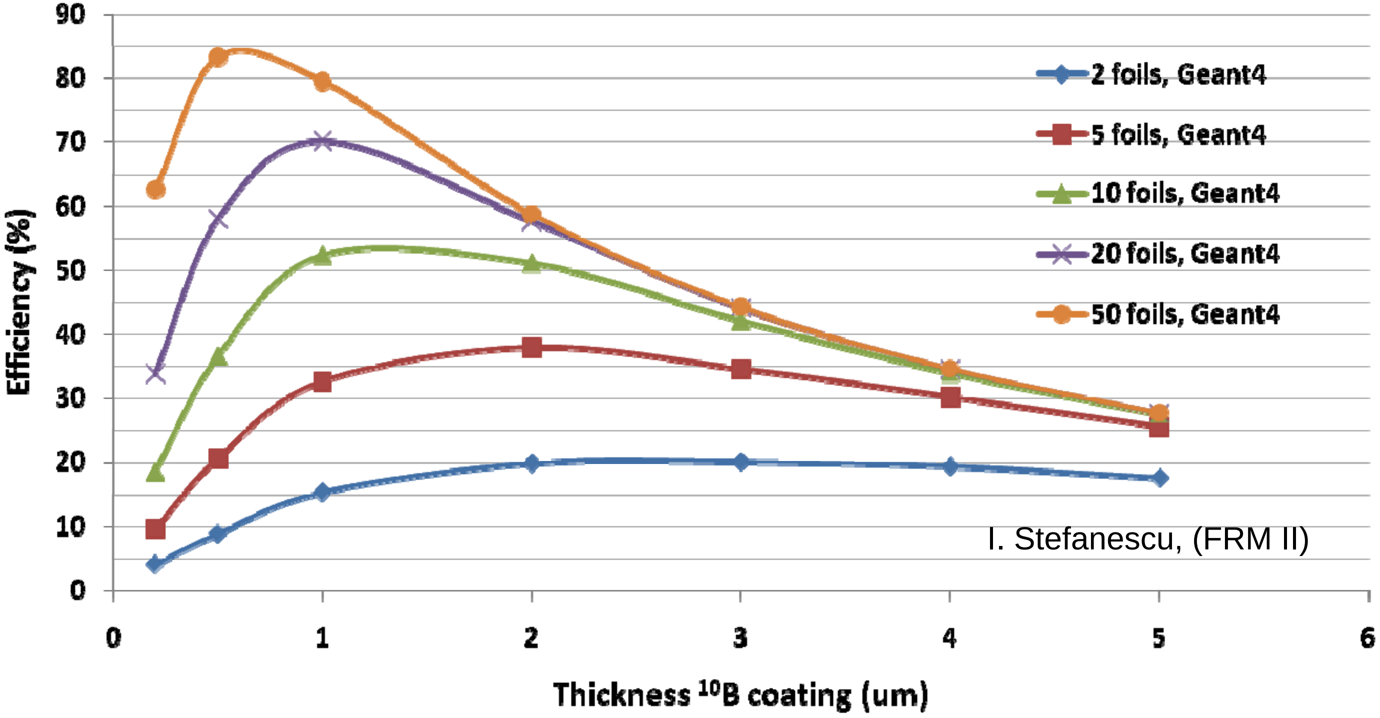
Detection efficiency of ^{10}B converters

Charged Particle range limits single layer efficiency

- Single layer: $\varepsilon_{det} < 5\%$ for therm. neutrons

➔ 20 -30 layers required for adequate efficiency !



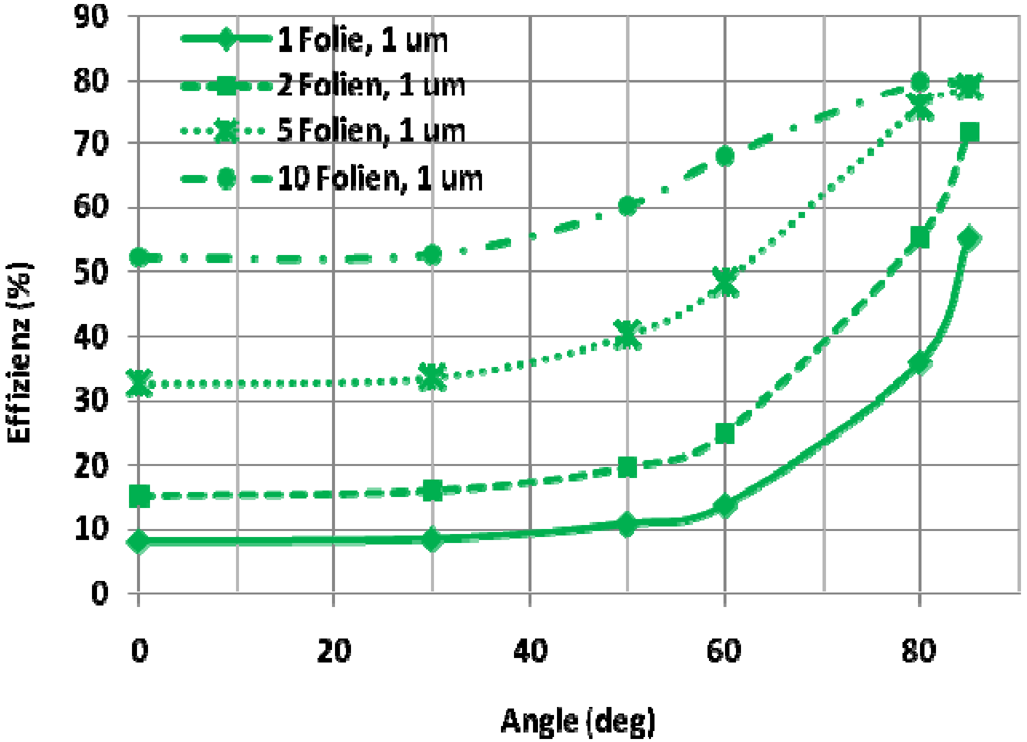


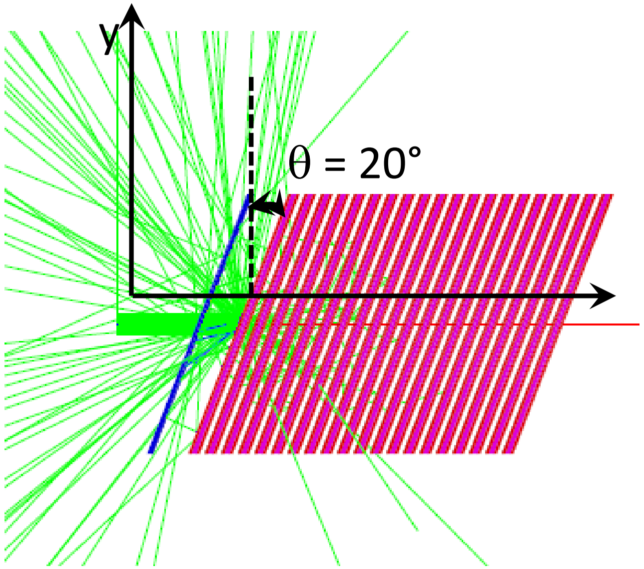
I. Stefanescu, (FRM II)

The optimum layer thickness decreases as the number of layers increases.

Detection efficiency of ^{10}B converters

Single layer efficiency could be increased by using an inclined geometry





I. Stefanescu, (FRM II)

Difficult to realize in a general purpose detector
Perhaps adequate to specific designs

^{10}B Multilayer detector concepts for large area detectors

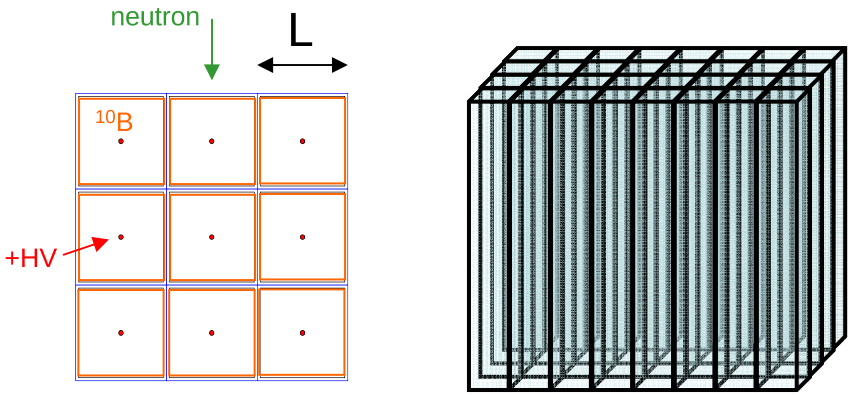
Modular multi-cell structure

- Different approaches & designs

  ILL: *“MultiGrid”*

  HZB: *“Microstructure profile”*

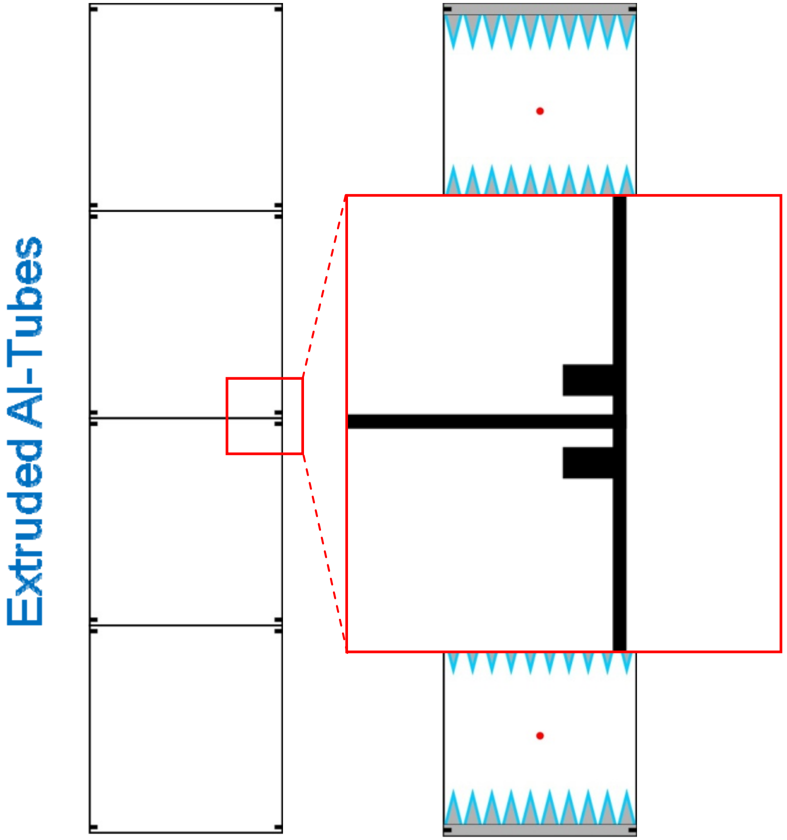
  Other designs ?



Present status:

A 16cm x 16cm prototype built at ILL has been successfully tested. An efficiency of 50% has been achieved with 28 layers of B_4C for 2.5 A neutrons.

Example: design proposed by HZB

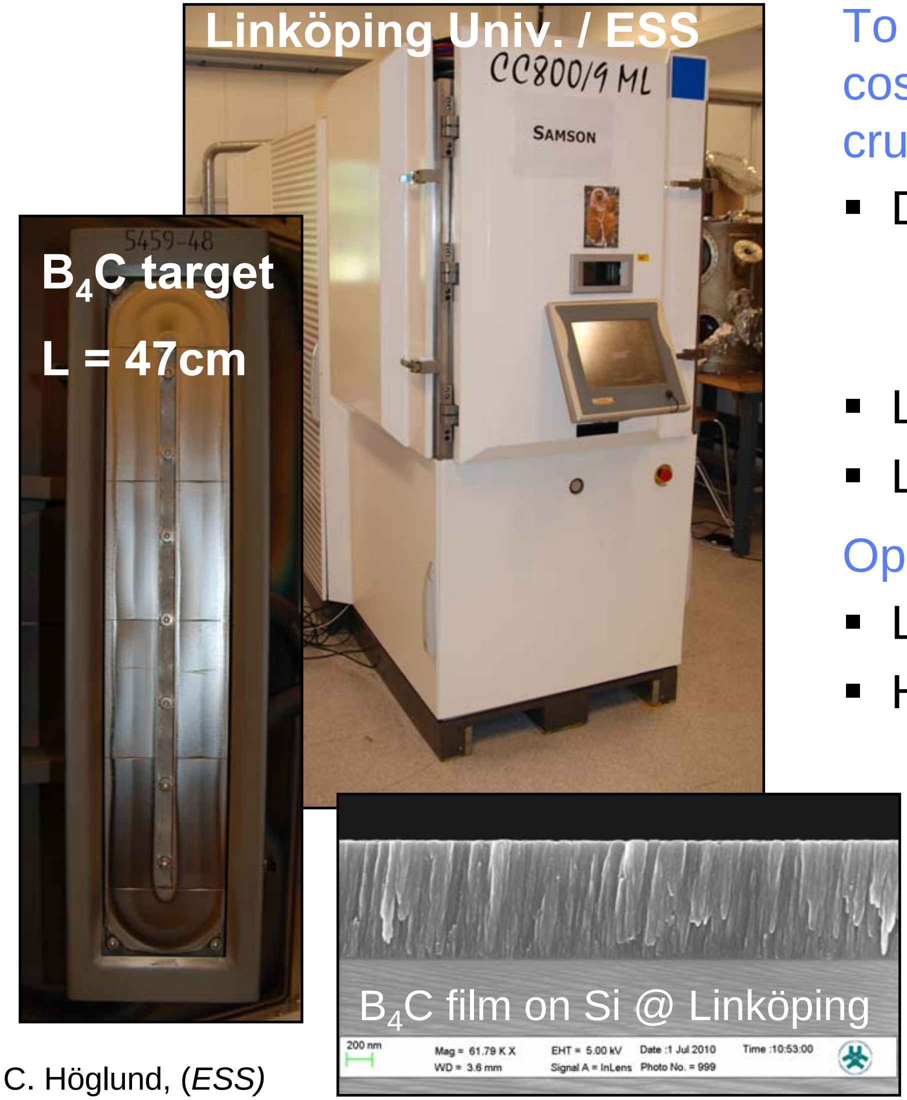


Short converters,
inserted in long tubes

Features are not to scale!

^{10}B / $^{10}B_4C$ layer production

DC Magnetron sputtering facility


Linköping Univ. / ESS

B_4C target

L = 47cm

B_4C film on Si @ Linköping

C. Höglund, (*ESS)*

To use this technology in large area detectors a cost effective production of the Boron layers is of crucial importance

- Deposition technologies

  RF / DC sputtering, e-beam evaporation, others

- Large scale production (~10^3 m^2)
- Layer composition: ^{10}B, $^{10}B_4C$, …

Open questions

- Layer stability: adhesion, ageing
- Homogeneity, substrate, topology

At Linköping Univ. meanwhile about 1400 “Al-blades” have been successfully coated on both sides with $^{10}B_4C$ to be used in a 200cm x 8cm prototype detector built at ILL

Present scintillation detector technology for neutron scattering

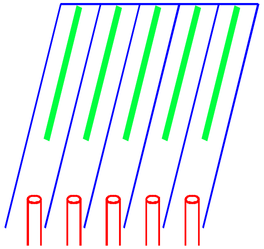
ZnS:^{6}LiF(Ag) scintillator

- read out by coded arrays of clear fibres

🟩 **SCINTILLATOR**

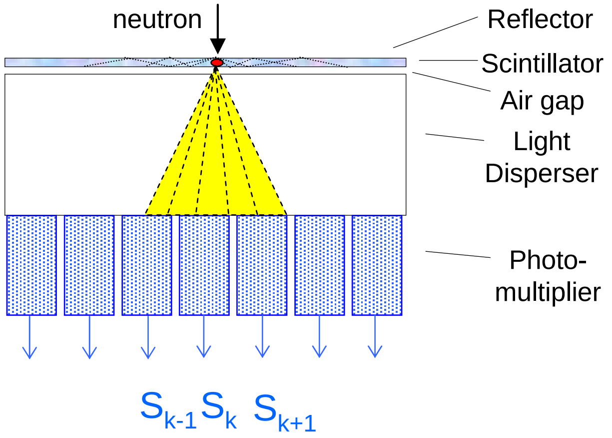
🟥 **FIBRE**

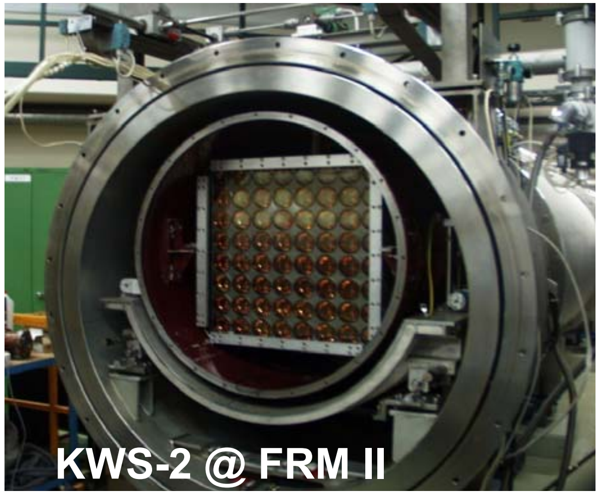
🟦 **REFLECTOR**



GS20 ^{6}Li-glass(Ce) scintillator

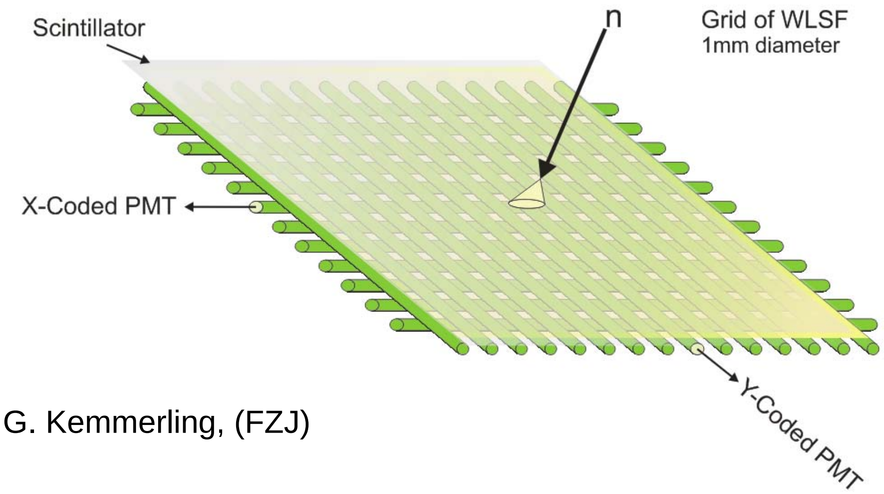
- read out by "Anger camera" array of photomultiplier tubes





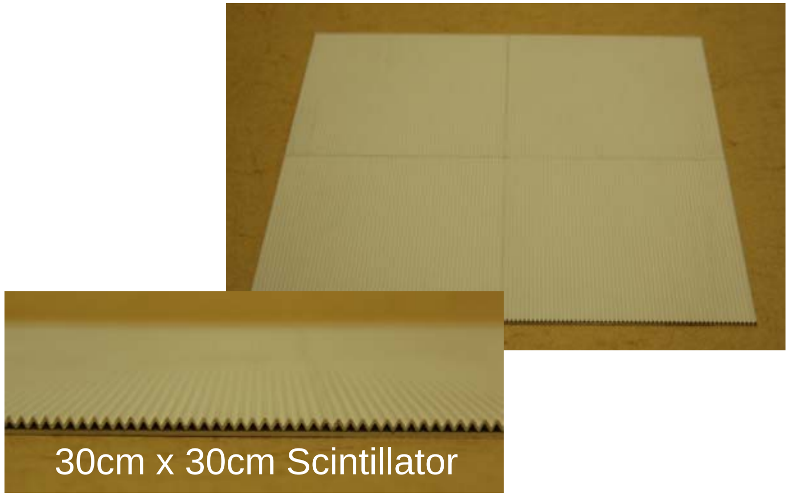
Wavelength Shifting Fibre Detector for Neutron Scattering

WLS-fibre readout of ^{6}LiF/ZnS & ^{10}B$_2$O$_3$ /ZnS-scintillators



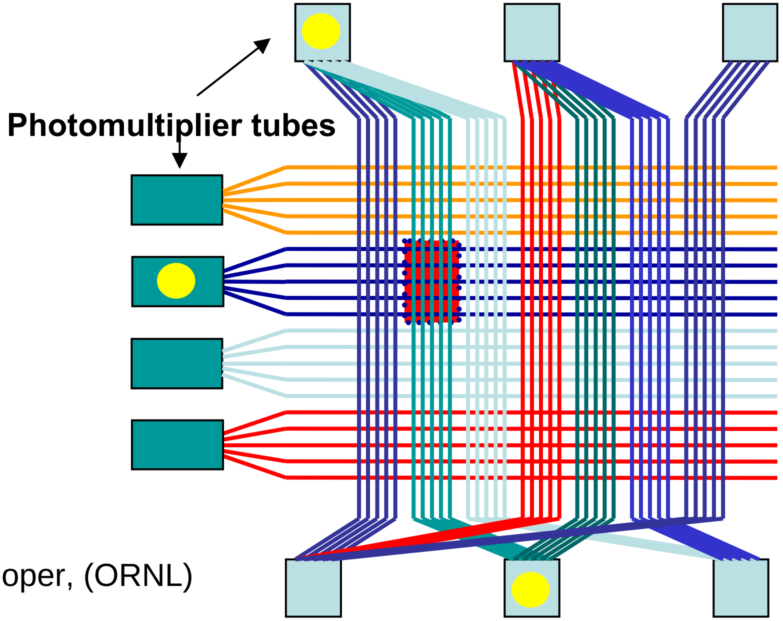
G. Kemmerling, (FZJ)

^{6}LiF/ZnS



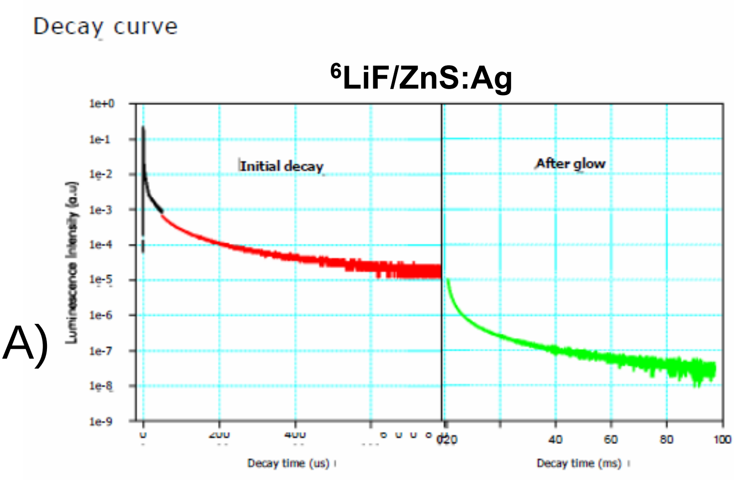
30cm x 30cm Scintillator

- **The incident neutron is captured in the ^{6}LiF/ZnS:Ag scintillator**
- **Some blue scintillation light from ZnS is shifted to green and trapped in the WLS-fibre**
- **This light is detected by PMTs in coincidence to determine the position**



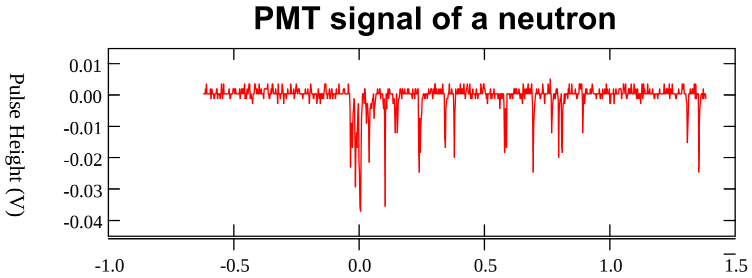
R. Cooper, (ORNL)

Challenges to a large area WLSF - detector



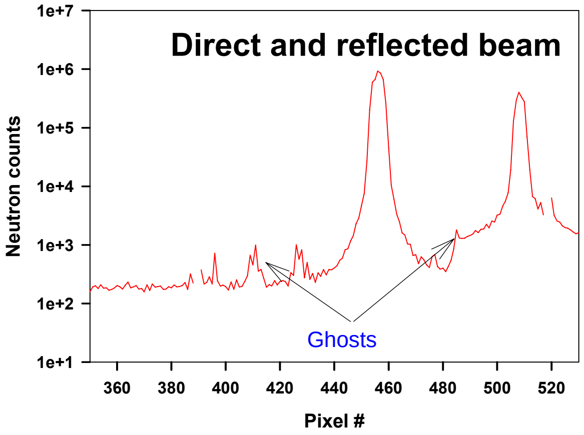
^{6}LiF/ZnS:Ag is a bright but slow scintillator

- decrease to 10% level is 80µs; "afterglow"
- limits local count rate capability to ~ 20kHz
- opaqueness limits neutron efficiency (50% @1.8A)



Low light output of WLS-fibres

- low light conversion and trapping efficiency
- losses due to damping and fibre bending



"Ghosting" (misplacement of neutrons)

- Occurs when afterglow from 2 neutron events cause signals in PMTs

Graphs taken from

G. Kemmerling, (FZJ); R. Cooper. (ORNL); E. Schooneveld, (*ISIS*)

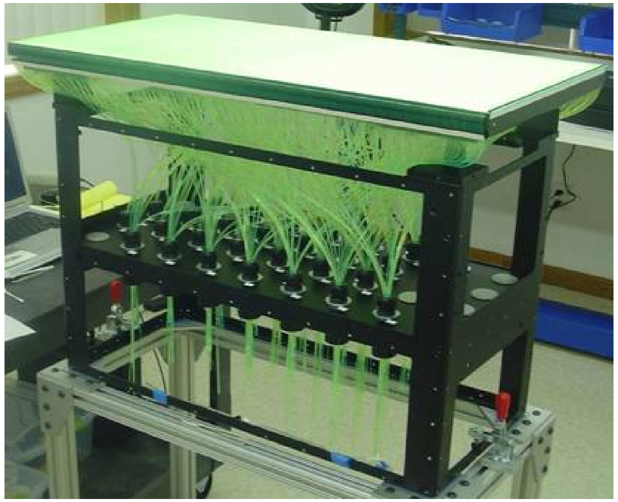
WLSF prototype for reflectometers

- Linear WLSF
- 768 elements
- DB16 coding: 32 PMTs for 128 elements
- 2 Flat ZnS sheets
- 768 fibres, 0.51mm pitch 0.5mm ∅, MS(300)
- 16 Channel MA PMT (H6568)
- New electronics: “Preamp” + discriminator
- Electronics on board

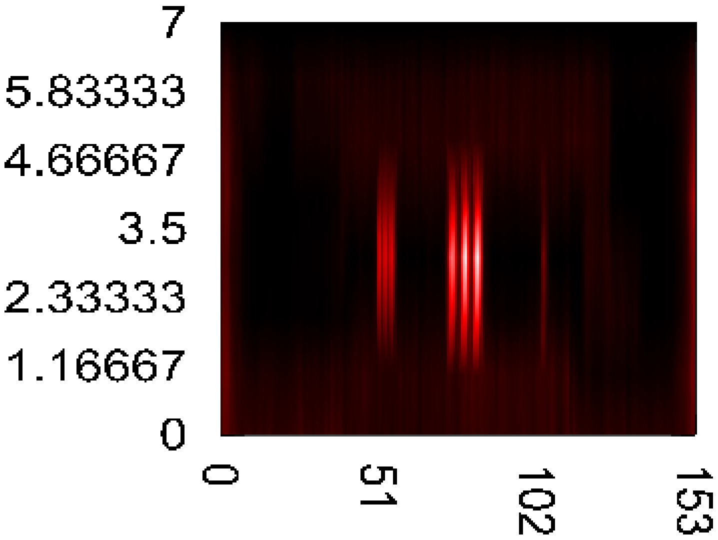
Science & Technology Facilities Council
ISIS

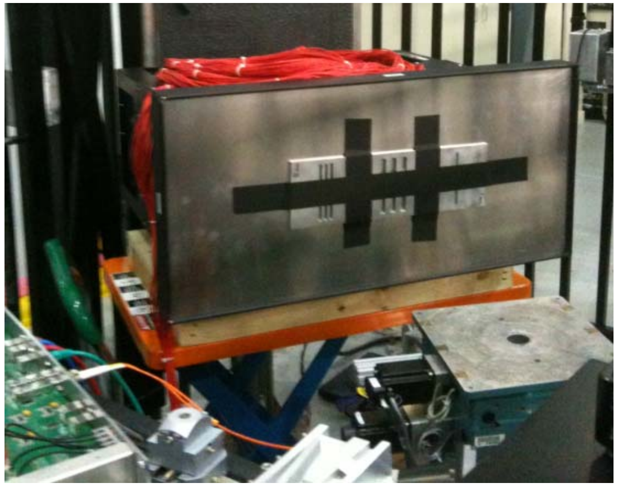
Fiber-scintillator detectors

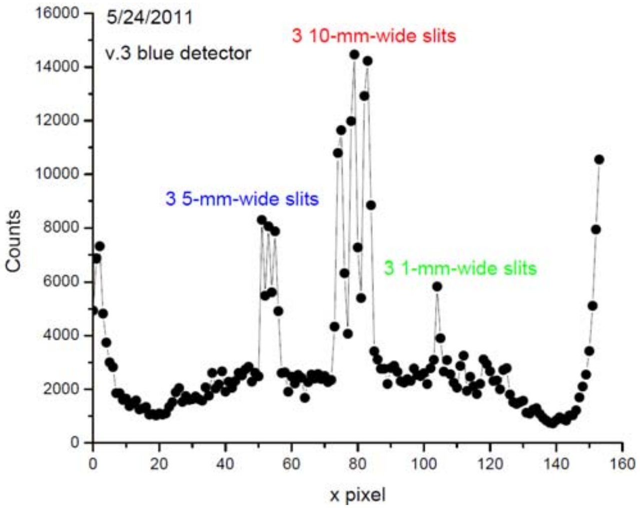
- Each module has 0.3 m^2 detection area.
- Totally 30 units (9 m^2) have been installed in 2 neutron diffractometers at SNS.
- The detector efficiency is as good as He-3 tube detectors (25 mm diameter at 10 bar).
- It is cost-effective to replace He-3 detectors.



Spatial-resolution for detector with new encoding





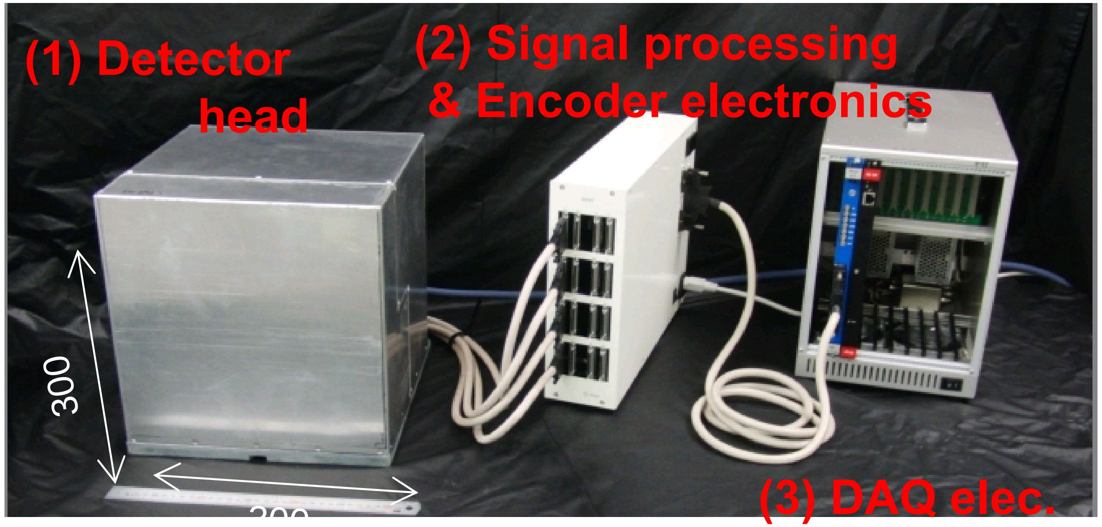


- Fiber mapping is a little different from the present version.
- Spatial resolution along x-axis is 4.1±0.2 mm.
- Ghosting (or artifact) is greatly reduced at high rate.
- Each pixel has 5 x 50 mm size, suitable for material studies in powder diffraction beamlines.

The new type of 2-d WLS fibre scintillator detector

A wide area scintillator detector has been developed using the iBIX detector technology.

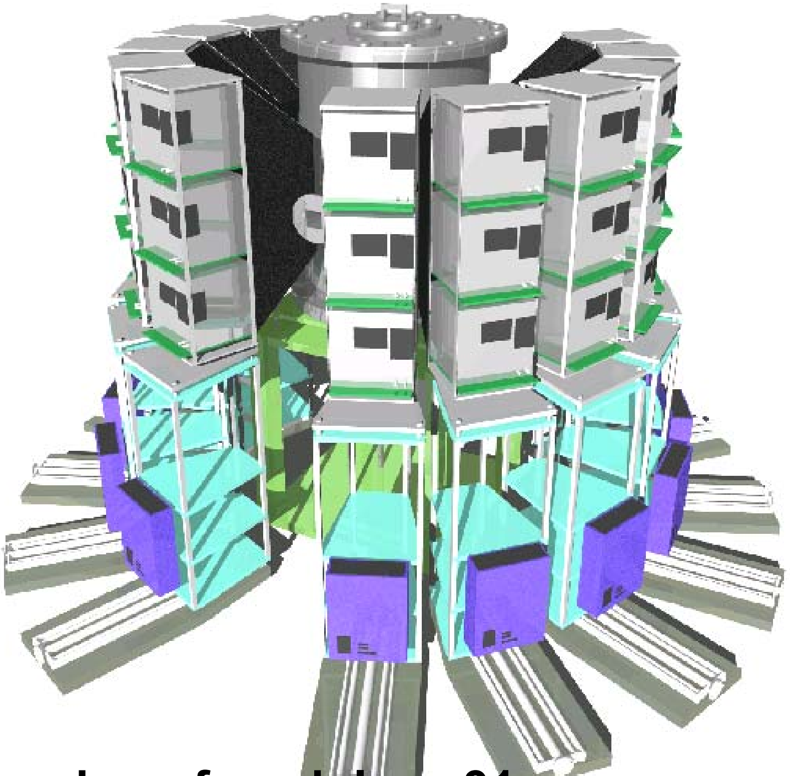
Detector module



Pixel size : 4 × 4 mm
Sensitive area : 256 x 256 mm
Detector efficiency : 30-40% for 1.8Å
Pulse pair resolution : < 5 μs
Gamma sensitivity : ~10^{-6} (^{60}Co)
No. of pixels /detector : 4096 (64 ×64)
No. of electronics channels : 128 (64 x 2)
No. of PMTs : 2

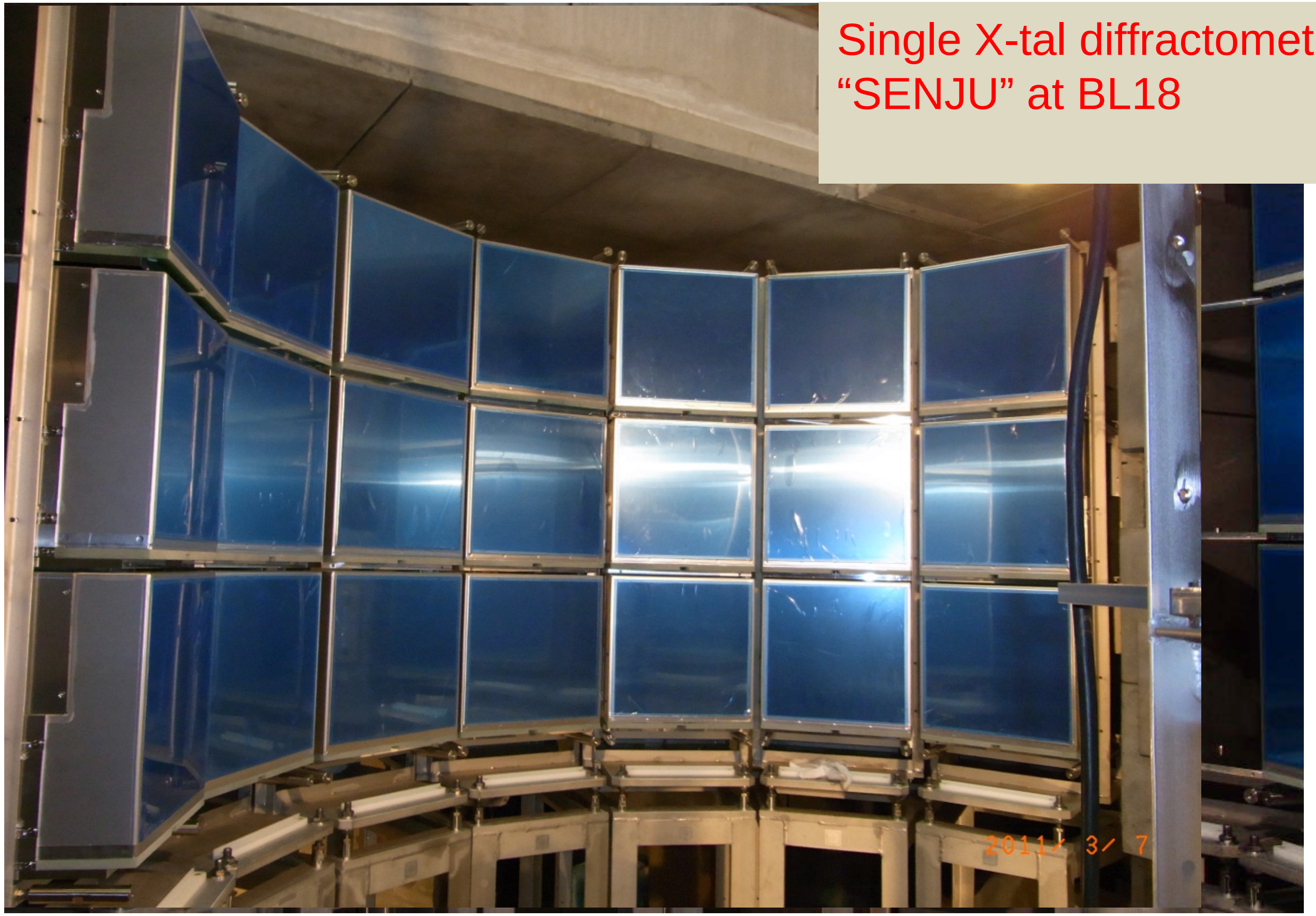
Single X-tal diffractometer "SENJU" at BL18

31 modules were fabricated, evaluated and installed.

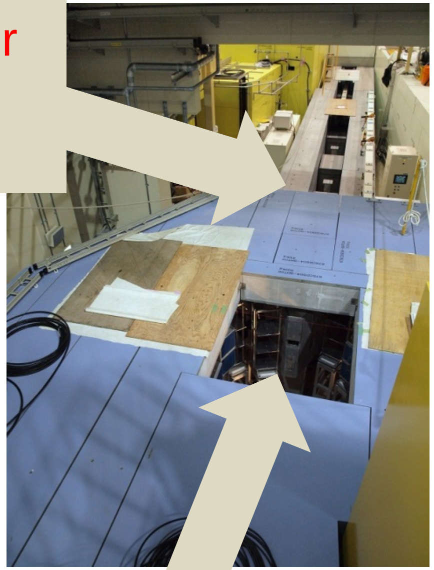


Number of modules : 31
Detection area : 2.8 m^2
Pixel size : 4 ×4 mm

Installation of “SENJU” Detectors at BL18



Single X-tal diffractometer
“SENJU” at BL18



Scintillator detectors
with WLSF read-out

31 detector modules
installed in the radiation shielding room of SENJU (BL18)

Conclusion and Outlook

As a reaction to the ongoing ^{3}He supply shortage a joint detector initiative formed by 9 international neutron scattering facilities has been formed

Three working groups evaluate alternative n-detection techniques based on

- ^{6}LiF/ZnS and B_2O_3/ZnS scintillation detectors with WLS-fibre readout
- Solid ^{10}Boron converter in gaseous detectors
- BF_3-filled Linear Position Sensitive Proportional Detectors

Prototype detectors are being build and show first promising results

There are still many open questions and problems to solve in order to achieve adequate performance of the new technologies and replace commonly used ^{3}He-detectors.

- Efficiency
- Count rate capability
- Cost issues

Helium-3 - a brief reminder

Helium-3 is a rare stable isotope with important applications:

- Cryogenics / Low temperature physics <1°K
- Medical lung imaging in conjunction with MRI
- Laser & other research
  - Helium-3 / Neon laser research
  - Gyroscopes (missile guidance & physics research)
- Neutron polarization (Helium-3 spin filter cells)
- **Neutron Detection**

5328b thermal neutron capture x-section + high pressure operation
➔ high efficiency, good γ / n separation < 10^{-6}, inert

mainly used for:

- US homeland security and non-proliferation programmes
- Neutron scattering applications
- Oil well logging and road construction

Helium-3 supply shortage

All available Helium-3 is a by-product of Tritium production for Nuclear Weapons Programs in the USA and Russia !

- Tritium decays via ß-decay into Helium-3 with a 12.3 years half-life
  Helium-3 separated and made available via DOE Isotope Program or Russia
- Tritium production reduced significantly due to disarmament
  US Tritium production stopped in 1988, resumed on small scale in 2003
- Until 2001 He-3 production exceeded demands, Since 2001 increased demand depleted US stock-pile from 235,000L to 40,000 L by 2009 !
- Security programs and neutron research claim a 5-years demand of ~ 250,000 L !
- DOE stops deployment of Helium-3 in 2009 !

Present supply situation:

- small flow of He-3 resumed from DOE stock-pile (for US users only)
  600 \$/L for government use / fed. funded research; 1000 \$/L for commercial use
- Non-US users face uncertain supply and rocketing prices up to 2500 € -3500 € /L
- predicted supply of 8,000 L/Y from the US for the next 6 years
  presumably similar amount from Russia, situation non-transparent

Helium-3 a brief reminder

In nature Helium-3 occurs with low abundance in two main sources

- The atmosphere contains ~ 280 billion Liters of Helium-3
  He-concentration in air ~ 5ppm; ^{3}He / ^{4}He ratio ~ few ppm
- Natural gas reservoirs contain a He-concentration up to several %
  ^{3}He / ^{4}He ratio ~ 70 -200 ppb ; more promising

  Liquefaction and ^{3}He/^{4}He separation expensive, not used yet !

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