

# Alternative techniques to $^3\text{He}$ based neutron detectors for neutron scattering applications

K. Zeitelhack

for the

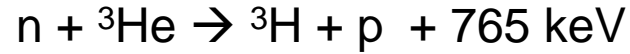
International Detector Initiative

# Outline

- Helium-3 demand and supply shortage
- Initiative to develop alternative techniques to Helium-3 based detectors
  - Scintillation detector technologies
  - Boron-10 converters in gaseous detectors
  - $\text{BF}_3$  – filled detector arrays
- Summary and Outlook

# Helium-3 use in neutron scattering

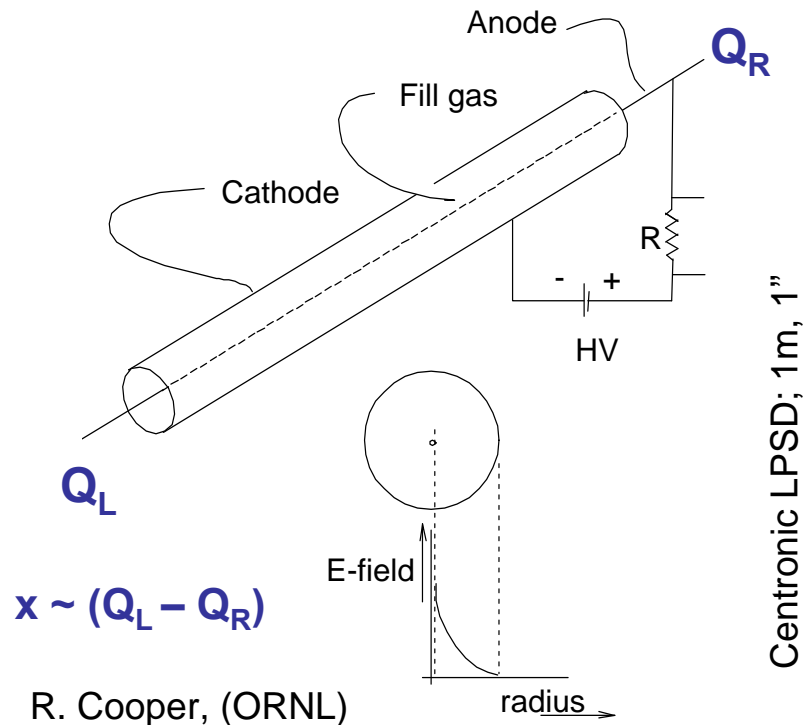
## Neutron Detection



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

~ 25,000 primary electrons / n

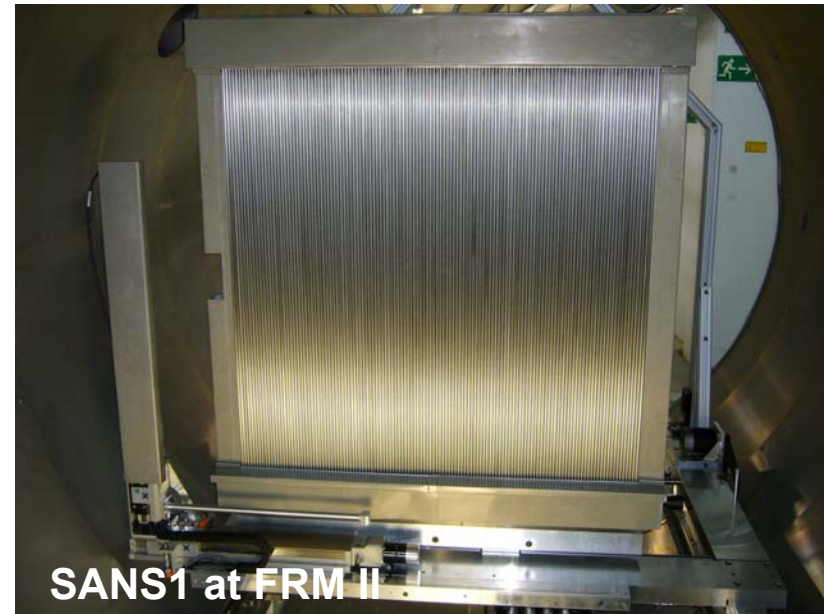
Gas Proportional Counter



Centronic LPSD; 1m, 1"

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

- highly efficient
- good position resolution
- stable
- low background
- very good n /  $\gamma$ -separation
- reasonable timing



SANS1 at FRM II

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

## Helium-3 Supply

### 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;  $^3\text{He} / ^4\text{He}$  ratio ~ few ppm
- Natural gas reservoirs contain a He-concentration up to several %  
 $^3\text{He} / ^4\text{He}$  ratio ~ 70 -200 ppb ; more promising

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

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

- Tritium decays via  $\beta$ -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 !

## 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*

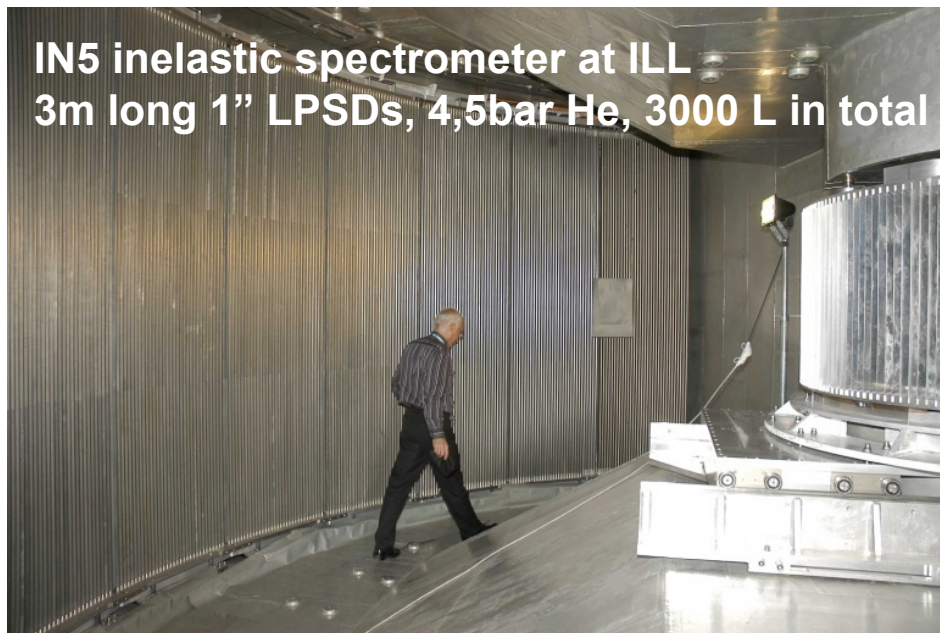
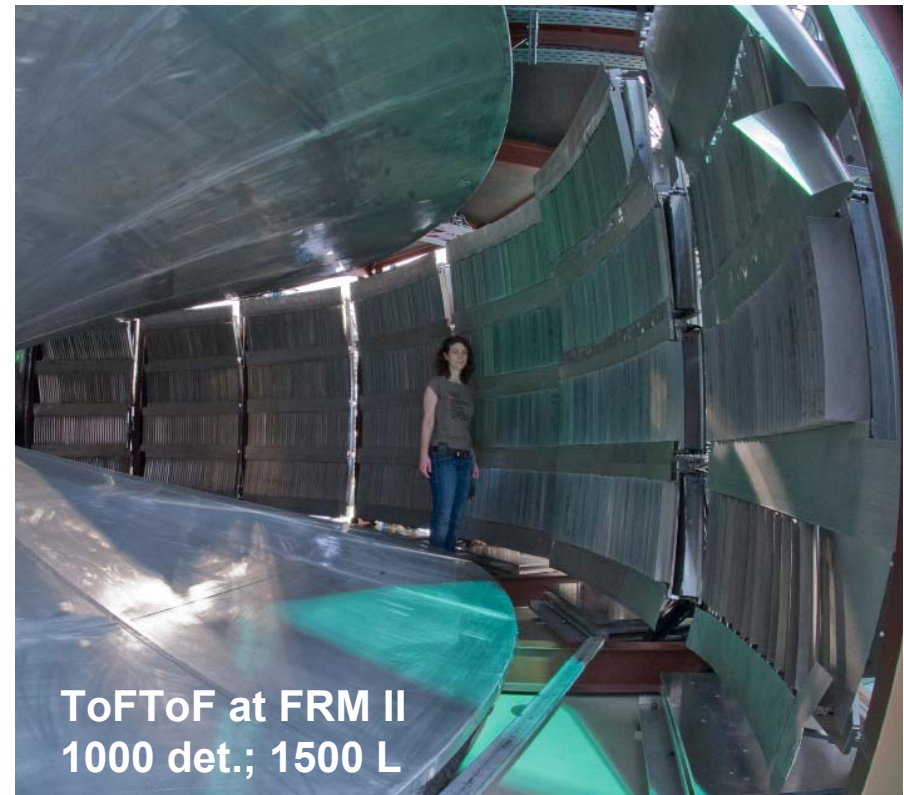
ORNL *Neutron Science Directorate, Oak Ridge National Laboratory, USA*

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





## Some Examples



- Typical Inelastic Instruments:**
- Detector area is 15 – 50 m<sup>2</sup>
  - 1" LPSDs, 2 – 3m long
  - He-3 content: 1000 - 4000 L

## Detector characteristics to compete

Detector characteristics	10 bar 25 mm diameter $^3\text{He}$
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 $\mu\text{s}$
Area	15 – 40 $\text{m}^2$
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
(✓)	$^3\text{He}$	gas	$^3\text{He}(n,p)t$	5333	7.59 bar-cm	P:573, t:191	$R_p = 0.43 \text{ bar-cm CF}_4$
(✓)	$^6\text{Li}$	solid	$^6\text{Li}(n,\alpha)t$	940	230 $\mu\text{m}$	T:2727, $\alpha$ :2055	$R_t = 130 \mu\text{m}$
✓	$^{10}\text{B}$	solid	$^{10}\text{B}(n,\alpha)^7\text{Li}$	3836	19.9 $\mu\text{m}$	$\alpha$ :1472, $^7\text{Li}$ :840	$R_\alpha = 3.14 \mu\text{m}$
✓	$^{10}\text{BF}_3$	gas	$^{10}\text{B}(n,\alpha)^7\text{Li}$	3836	9.82 bar-cm	$\alpha$ :1472, $^7\text{Li}$ :840	$R_\alpha = 0.42 \text{ bar-cm}$
✗	$\text{natGd}$	solid	$\text{natGd}(n,\gamma)$	49122	6.72 $\mu\text{m}$	Ce:29-182 (86.5%)	$\Lambda_{\text{ce}} = 12.3 \mu\text{m}$

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:<sup>6</sup>LiF(Ag) or ZnS:<sup>10</sup>B<sub>2</sub>O<sub>3</sub>(Ag) scintillators read out by coded arrays of clear or wavelength shifting fibres
- Investigate scintillators, optics, light readout devices, encoding schemes

### <sup>10</sup>B-Working Group (*ILL, ESS, FRM II, HZB, ORNL*)

#### Development of solid <sup>10</sup>Boron multilayer arrangements in gaseous large area neutron detectors

- Study <sup>10</sup>B-coating processes
- Investigate and optimize design and fabrication of a multilayer detector in view of performance and cost

### BF<sub>3</sub>-Working Group (*HZB, FRM II, ILL*)

#### Investigate BF<sub>3</sub> as a potential fast and easy replacement of <sup>3</sup>He

- Study gas properties, performance and limitations of BF<sub>3</sub>
- Investigate safety issues for large scale use

## $^{10}\text{BF}_3$ Proportional Counters

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

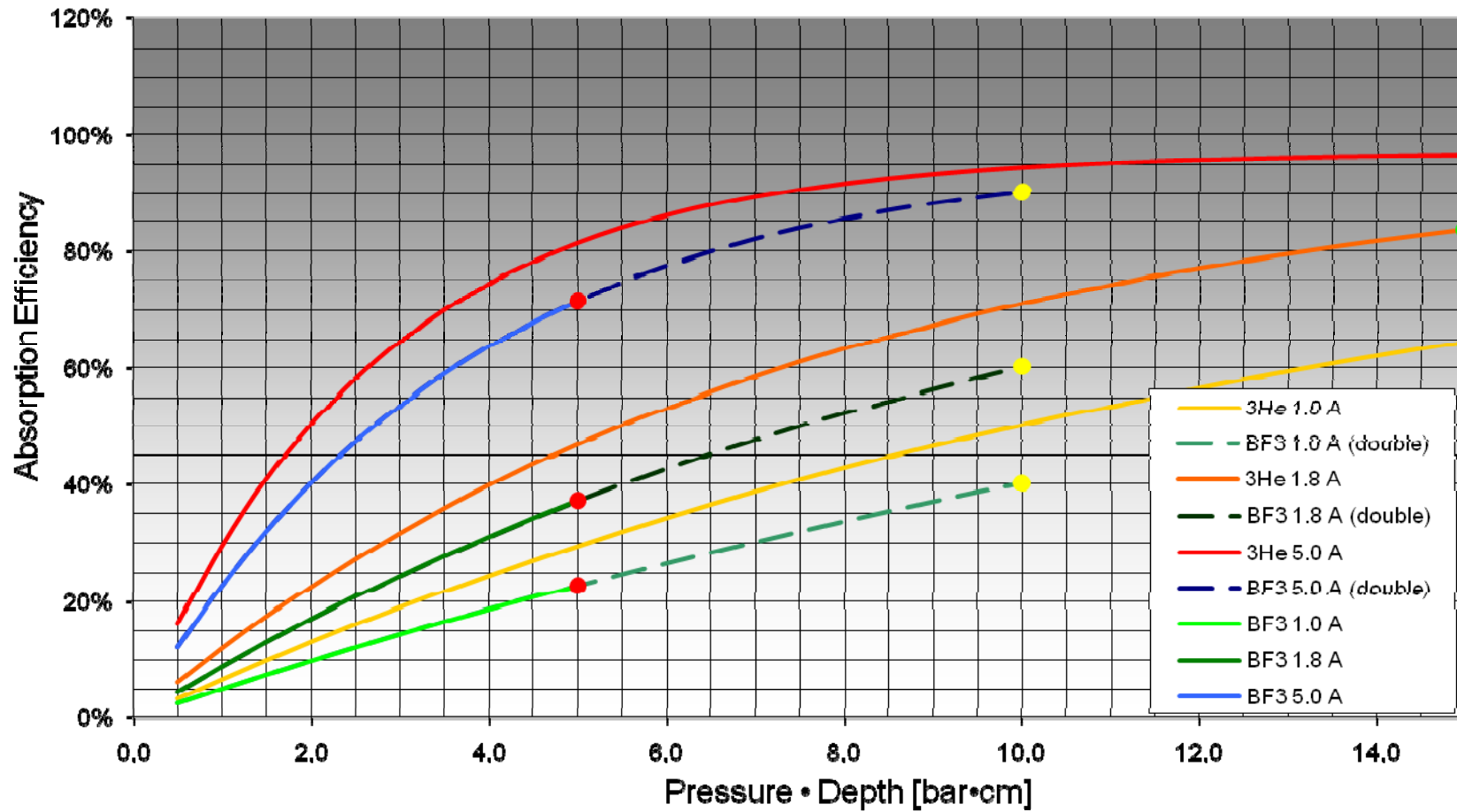
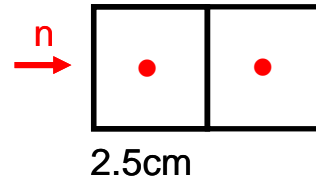
- Energy deposit 2.3 MeV / neutron
  - *large detector signals; good position resolution ( $\Delta L/L = 0.6\%$ )*
  - *excellent  $n / \gamma$ -separation ( $< 10^{-6}$ )*
- Cross section = 72% of Helium

### **BUT !**

- Efficiency limited by pressure (~2 bar) due to electron attachment in  $\text{BF}_3$ 
  - *several detector rows needed to achieve adequate efficiency depending on neutron wavelength*
- Corrosive and highly toxic
- High voltage increases rapidly with pressure

**→ Solution for cold neutrons only ?**

# Neutron absorption efficiency of $^{10}\text{BF}_3$

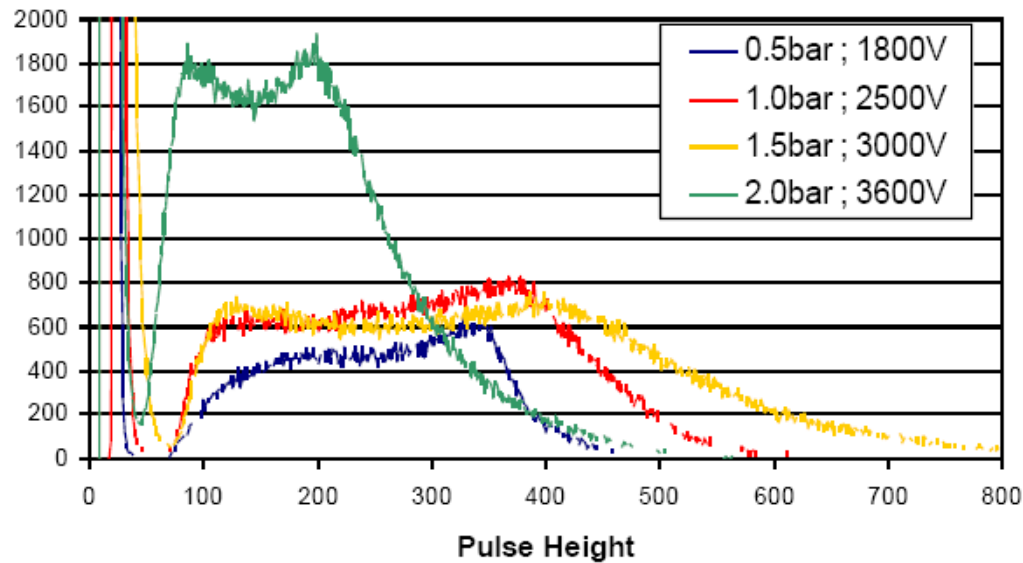


# LPSD filled with $^{10}\text{BF}_3$ at high pressure

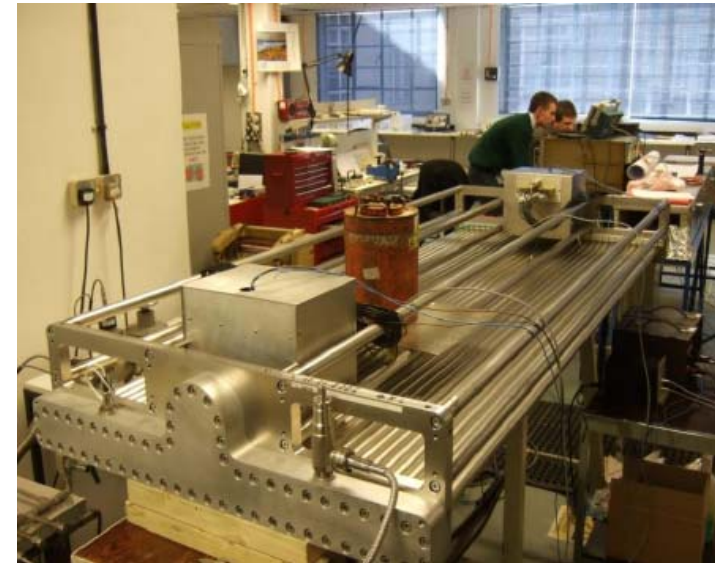
## Test of ILL- IN5 prototype module filled with $\text{BF}_3$

- 32 tubes; 1" x 2m
- $\text{BF}_3$  pressure up to 2bar

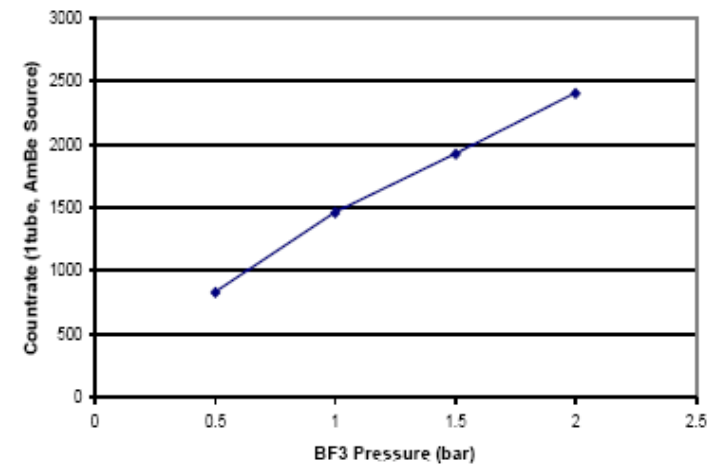
Spectra @ PSD Gain for various  $\text{BF}_3$  Pressures



B. Guerard, M. Platz, (ILL)



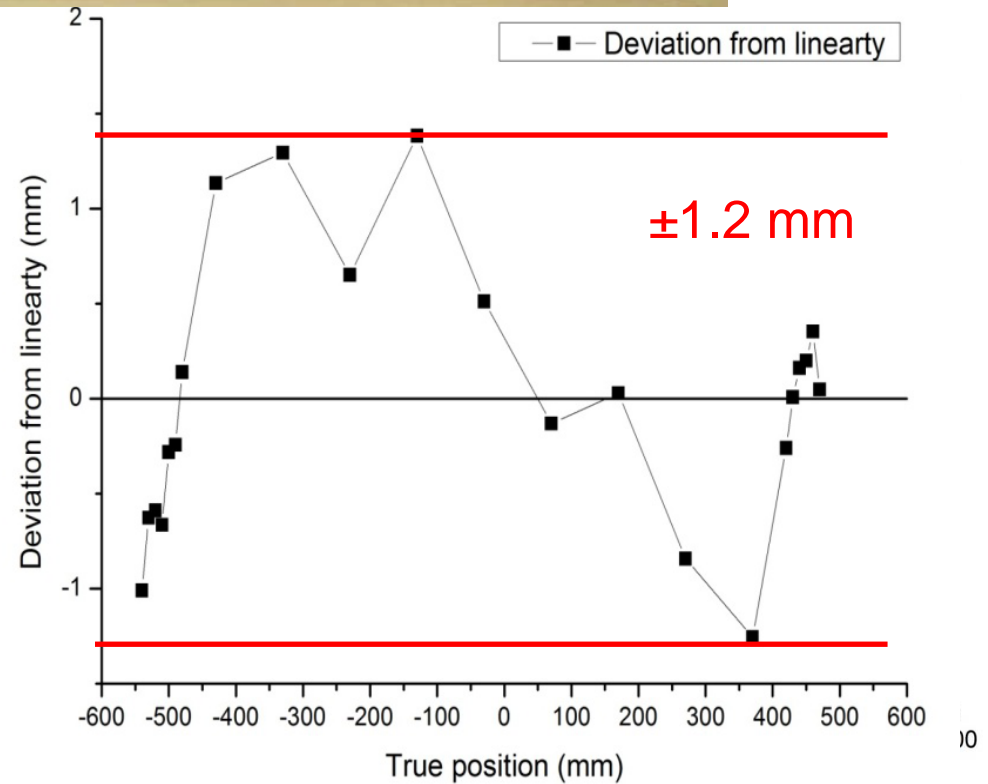
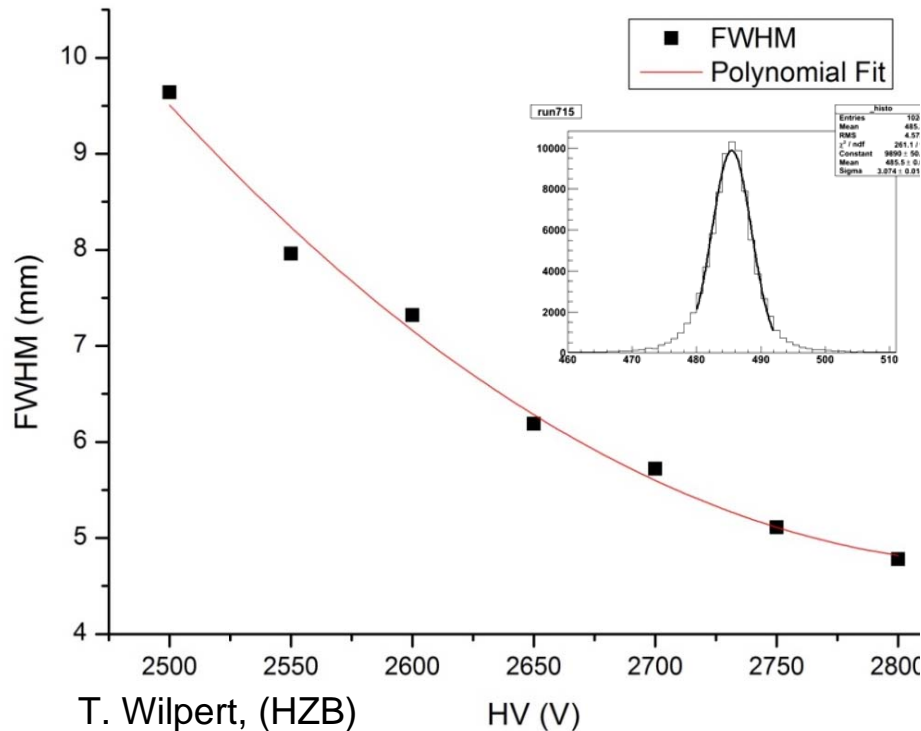
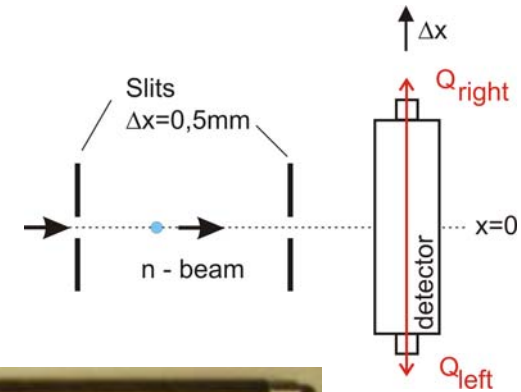
Count rate vs  $\text{BF}_3$  Pressure



# LPSD filled with $^{10}\text{BF}_3$ at high pressure

## Evaluation of PSD at FRM II and HZB

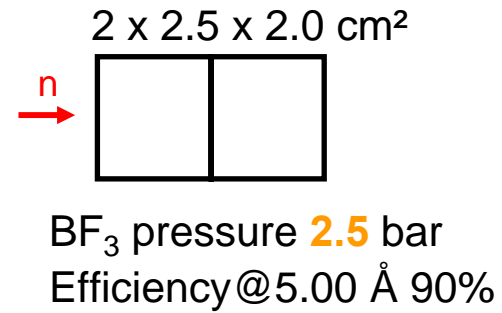
- 1" x 1m LPSD (made by Centronic, UK)
- 1.87 bar, 2800 V max. tested





# NEAT – ToF spectrometer at HZB

40 m<sup>2</sup> active detector area  
544 LPSDs (17 Modules)  
Pixel resolution 2.5 x 2.5 cm<sup>2</sup>  
ToF resolution < 30 μs @ 5Å  
1" width, 3 m long, 90% @ 5Å  
Radius of detection plane 3 m  
In-plane angle 280°

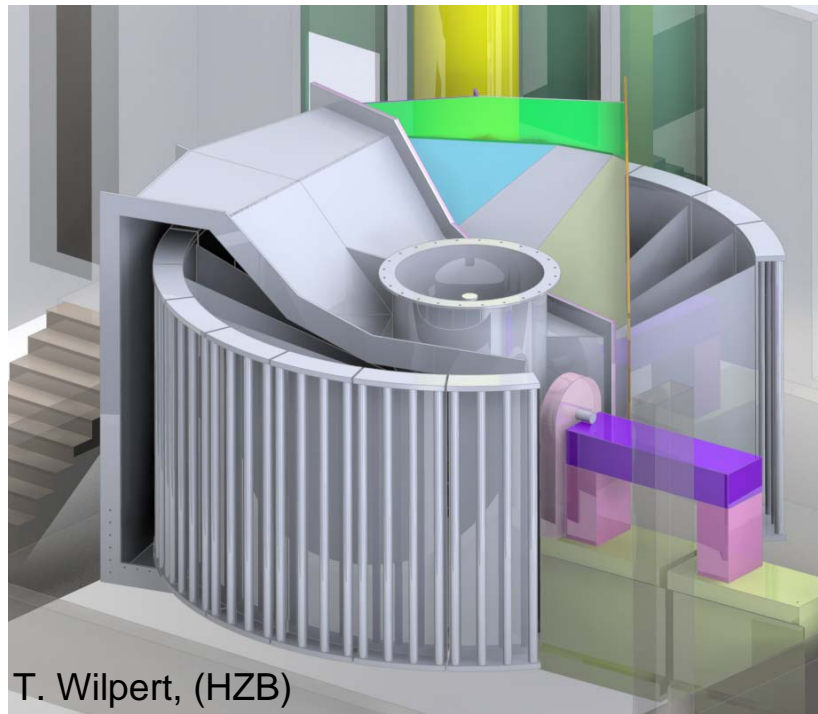


Detector module with 32 double-tubes

Backside

Front-side

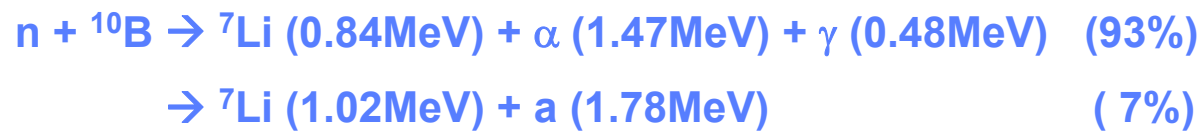
LN<sub>2</sub>-cold trap



# <sup>10</sup>B-converter in gaseous detectors

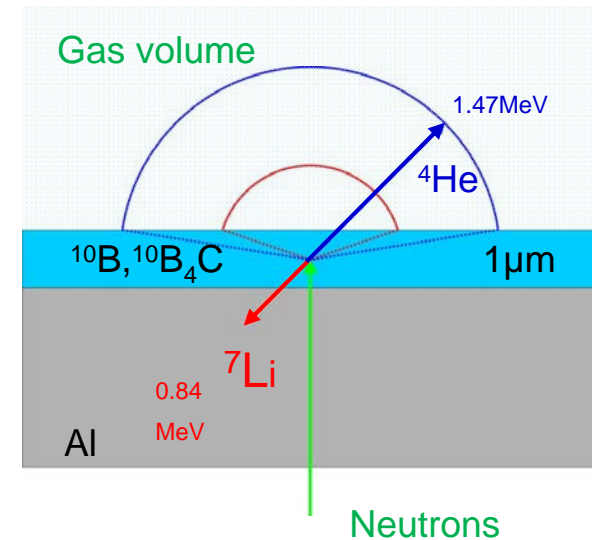
## Why using Boron as solid converter ?

- B<sub>4</sub>C stable, not hygroscopic (e.g. as Li)
- large charge signal in detector
- 96% enriched <sup>10</sup>B available



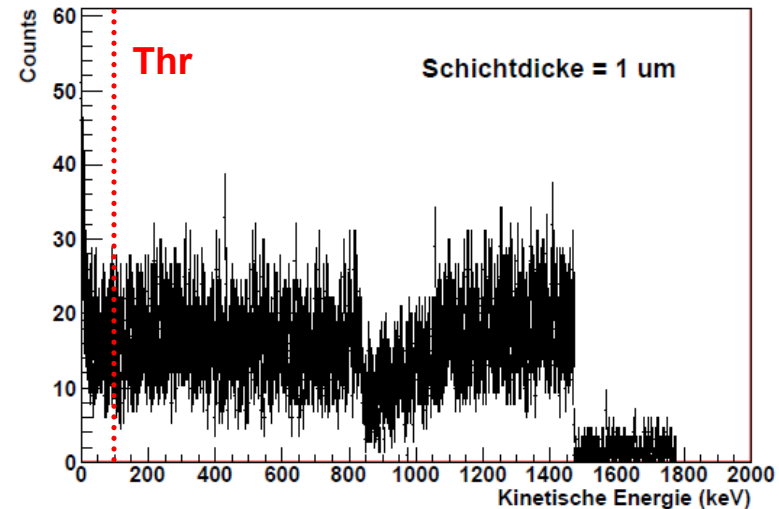
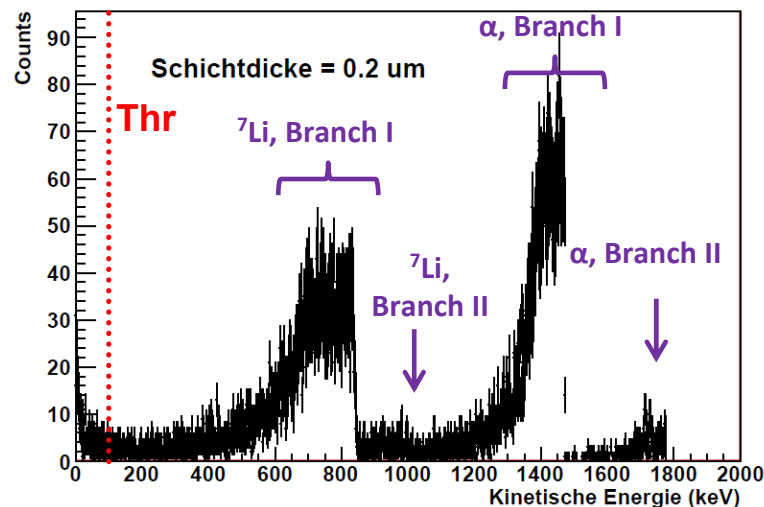
$\lambda_{\text{abs}}$  for therm. neutrons: 20  $\mu\text{m}$

Range in Boron:  $\alpha = 3.14 \mu\text{m}$ ; Li = 1,53  $\mu\text{m}$



I. Stefanescu, (FRM II)

Pulse height spectra simulated with GEANT4

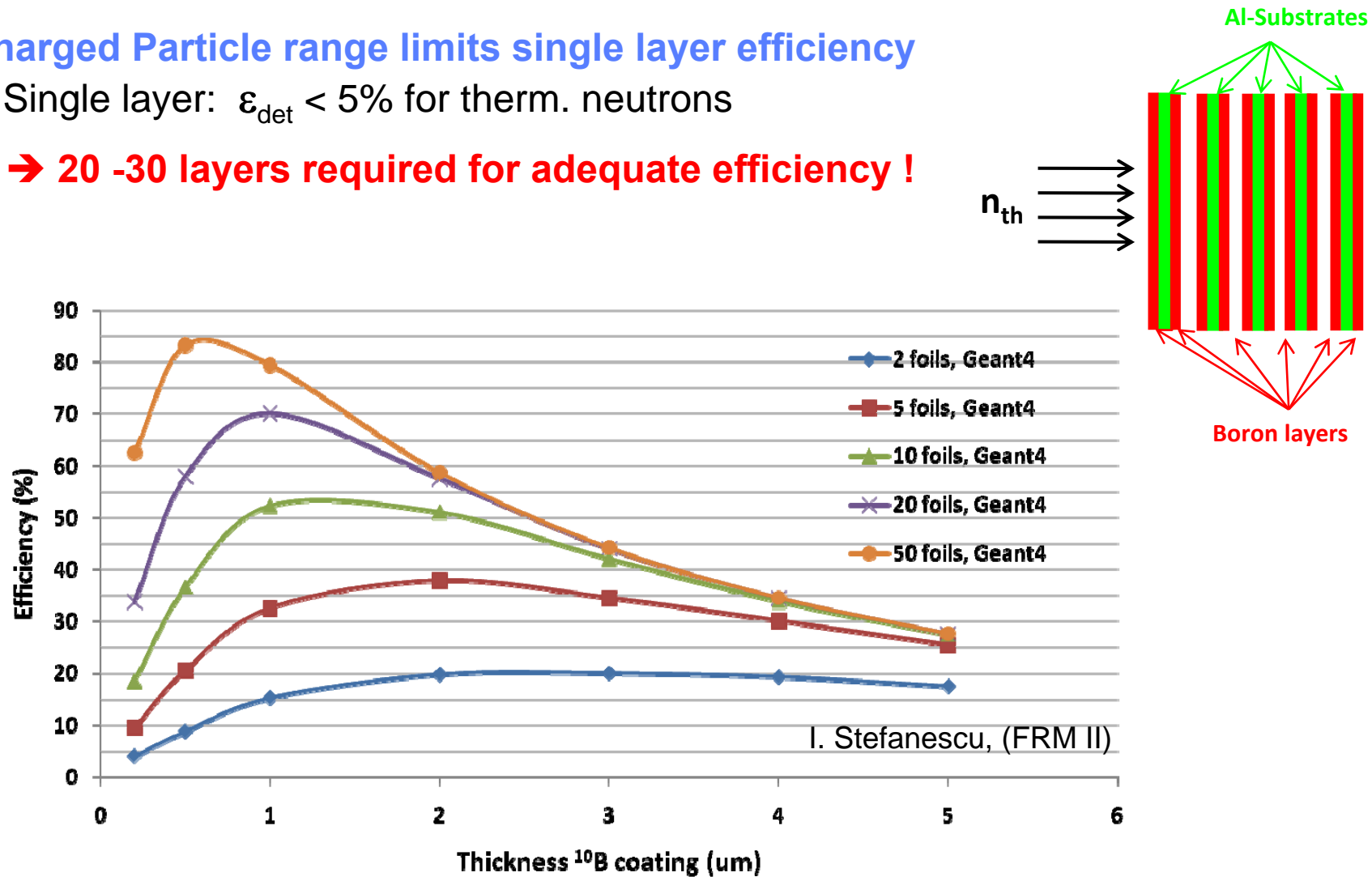


## Detection efficiency of $^{10}\text{B}$ converters

### Charged Particle range limits single layer efficiency

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

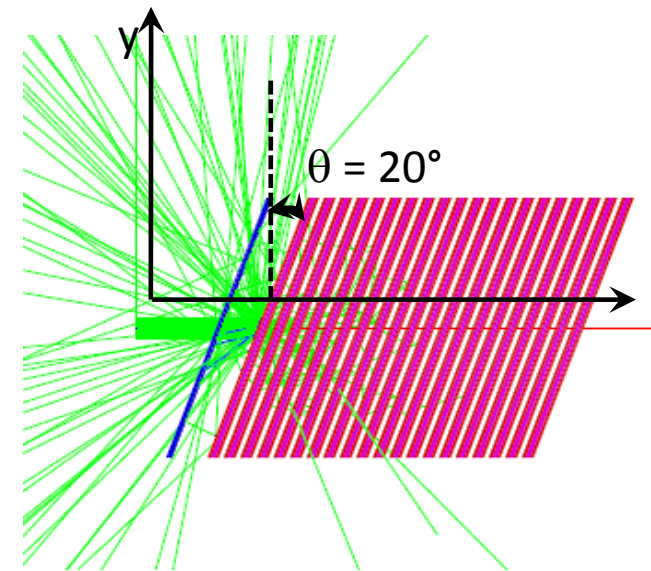
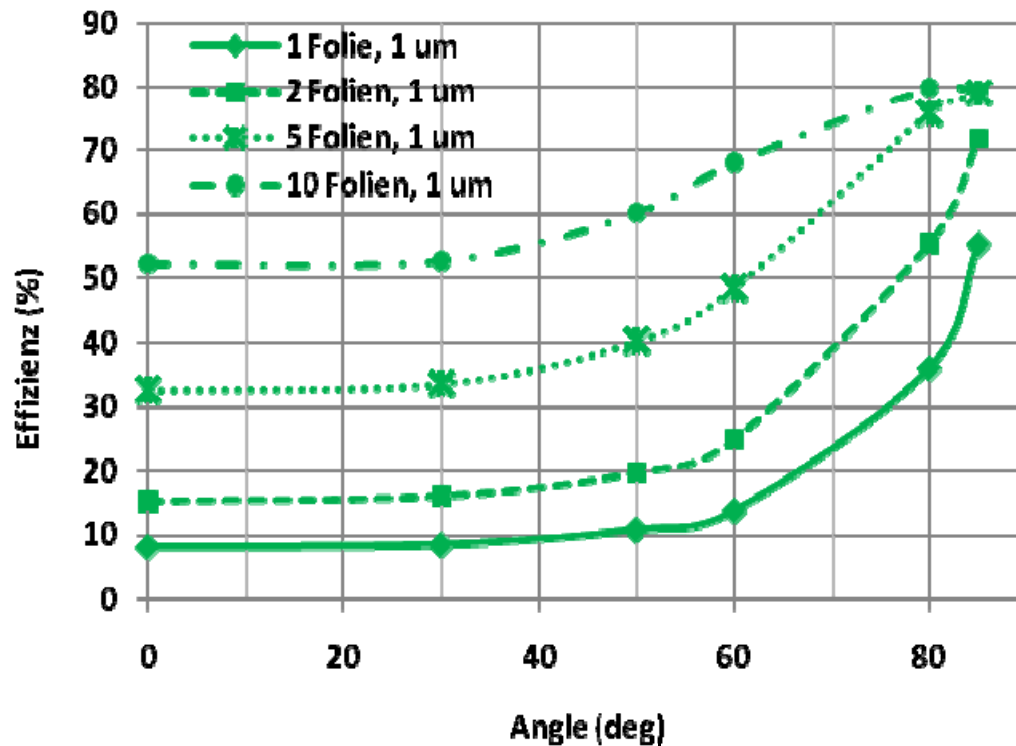
→ 20 -30 layers required for adequate efficiency !



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

# Detection efficiency of $^{10}\text{B}$ 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

# $^{10}\text{B}$ / $^{10}\text{B}_4\text{C}$ layer production

DC Magnetron sputtering facility



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 ( $\sim 10^3 \text{ m}^2$ )
- Layer composition:  $^{10}\text{B}$ ,  $^{10}\text{B}_4\text{C}$ , ...

## 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  $^{10}\text{B}_4\text{C}$  to be used in a 200cm x 8cm prototype detector built at ILL

C. Höglund, (ESS)



# $^{10}\text{B}$ Multilayer detector concepts for large area detectors

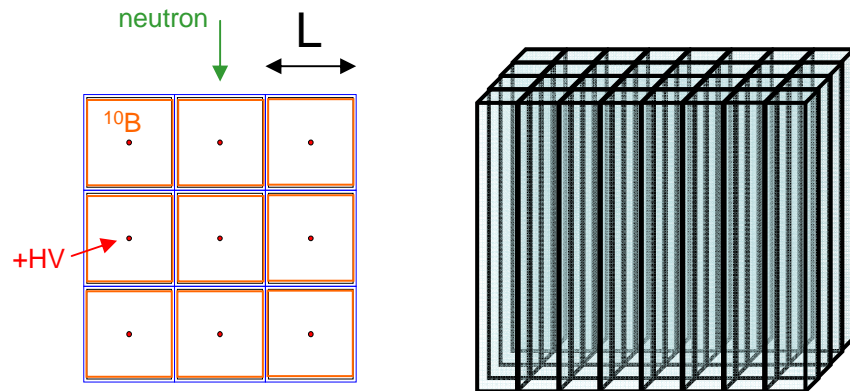
## Modular multi-cell structure

- Different approaches & designs

ILL: “MultiGrid”

HZB: “Microstructure profile”

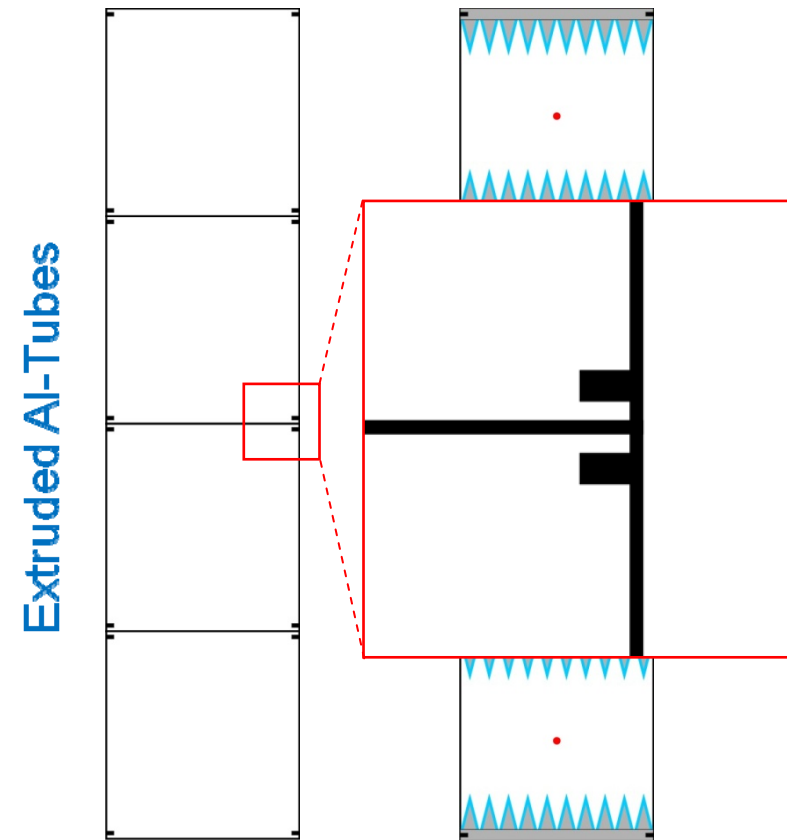
Other designs ?



## 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  $\text{B}_4\text{C}$  for 2.5 Å neutrons.

## Example: design proposed by HZB



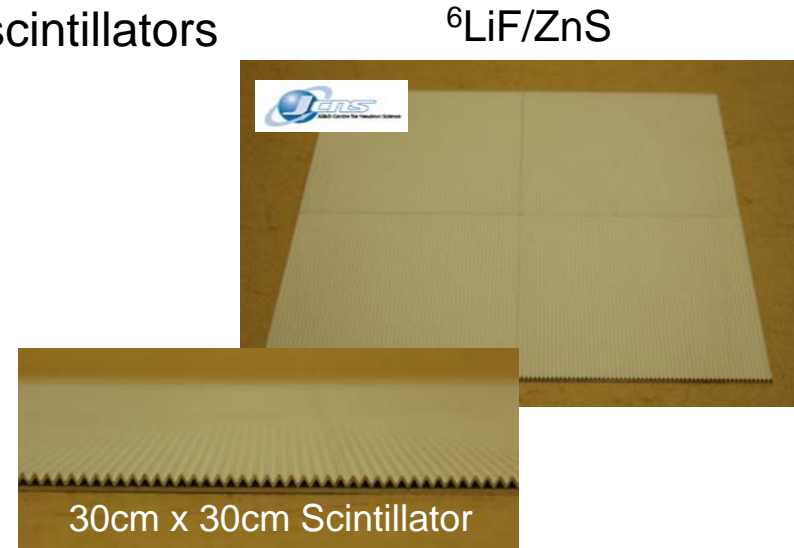
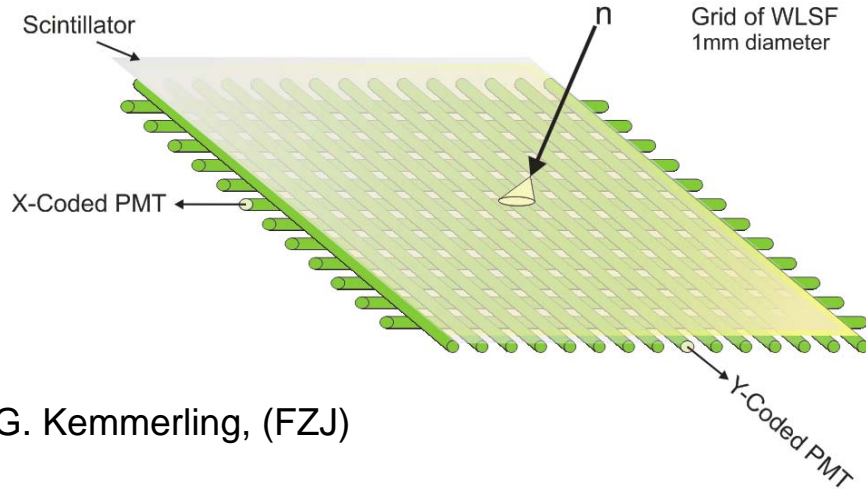
Short converters,  
inserted in long tubes

Features are not to scale!



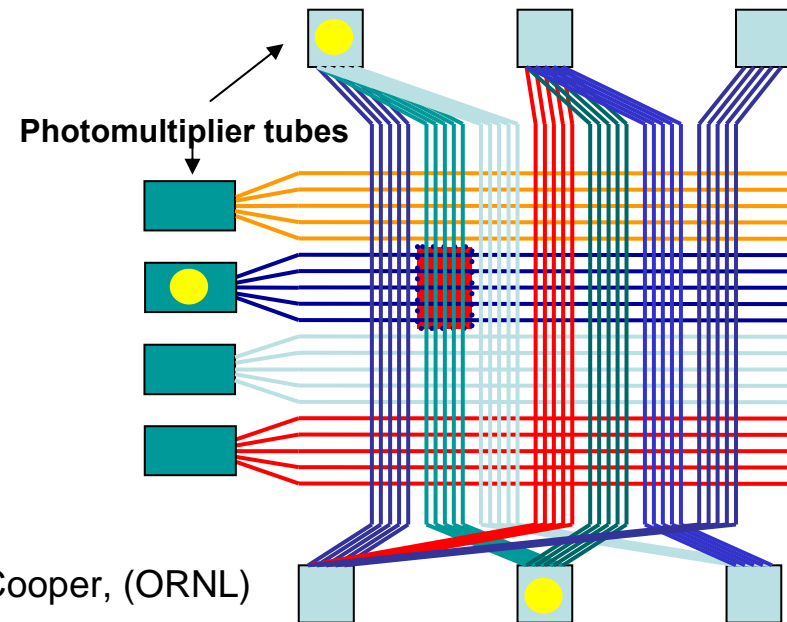
# Wavelength Shifting Fibre Detector for Neutron Scattering

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



G. Kemmerling, (FZJ)

- The incident neutron is captured in the  ${}^6\text{LiF/ZnS:Ag}$  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

## $^6\text{LiF/ZnS:Ag}$ is a bright but slow scintillator

- decrease to 10% level is  $80\mu\text{s}$ ; “afterglow”
- limits local count rate capability to  $\sim 20\text{kHz}$
- 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

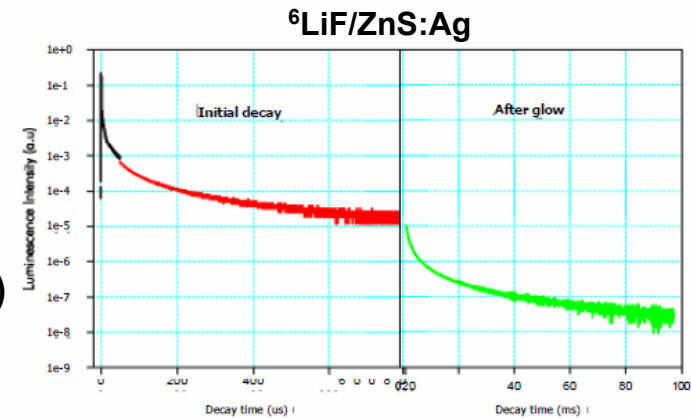
## “Ghosting” (misplacement of neutrons)

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

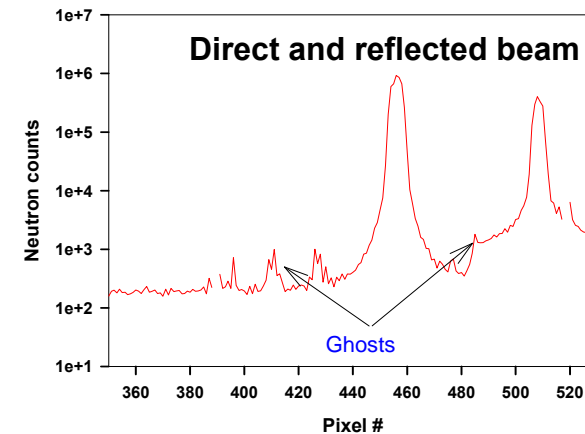
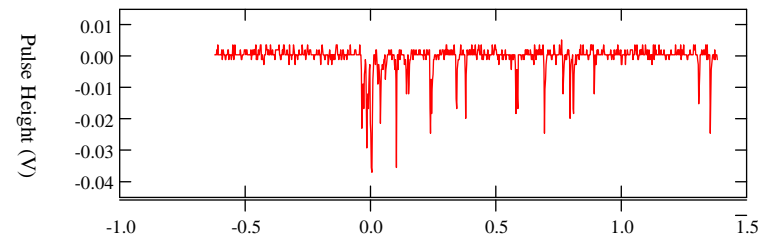
Graphs taken from

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

Decay curve



PMT signal of a neutron



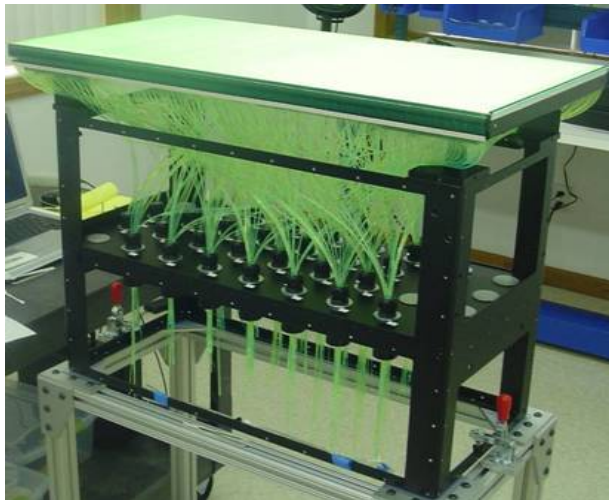
## WLSF prototype for reflectometers

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

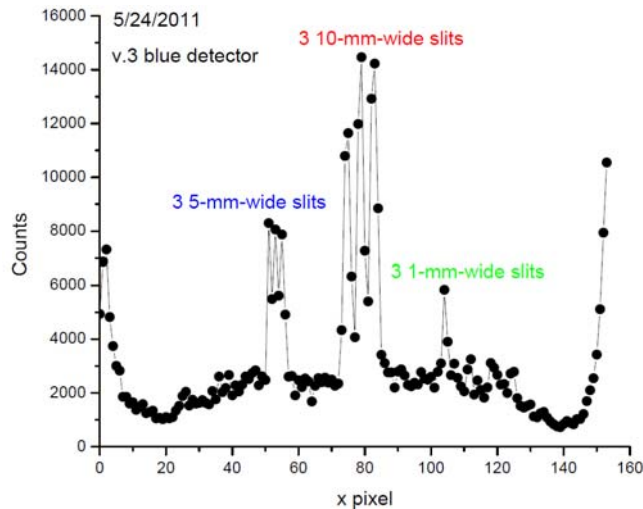
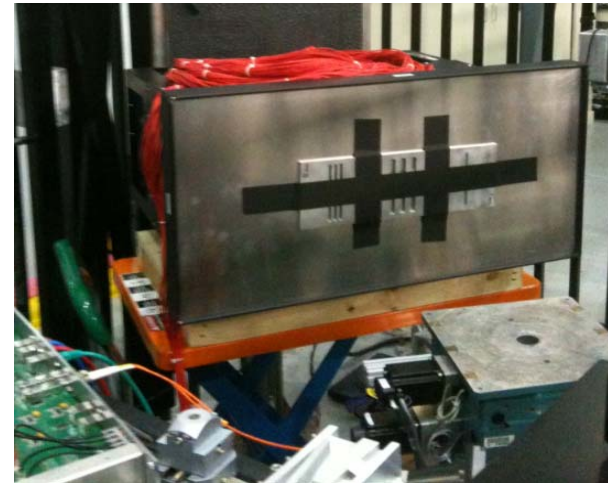
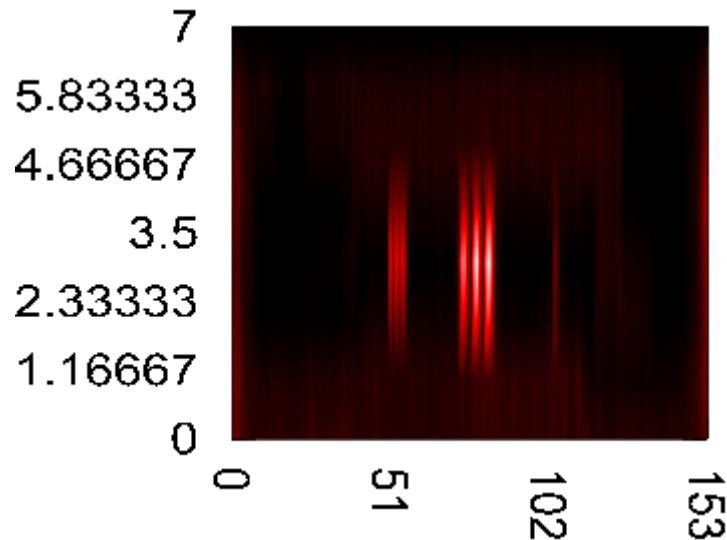


# Fiber-scintillator detectors

- Each module has 0.3 m<sup>2</sup> detection area.
- Totally 30 units (9 m<sup>2</sup>) 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  $4.1 \pm 0.2$  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.

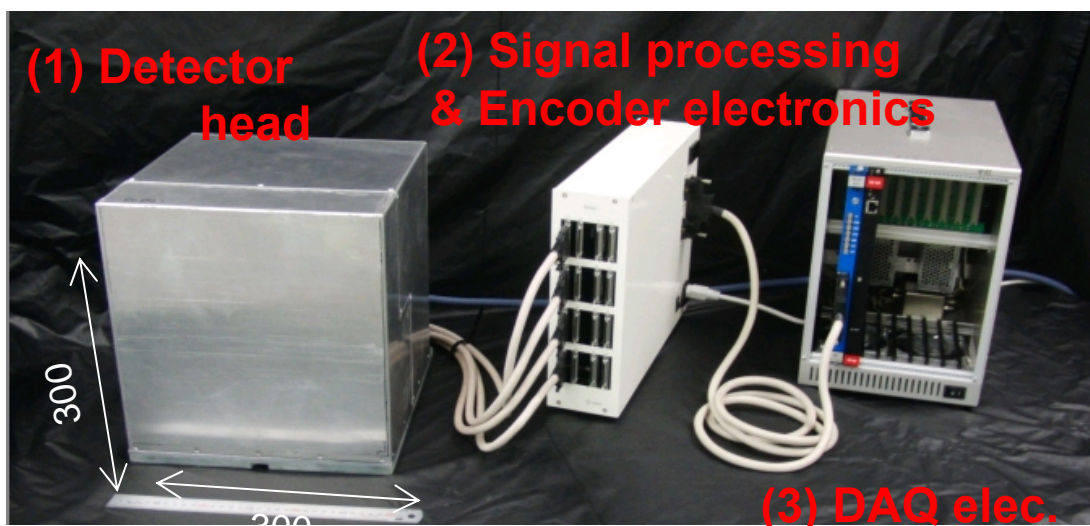


# The new type of 2-d WLS fibre scintillator detector

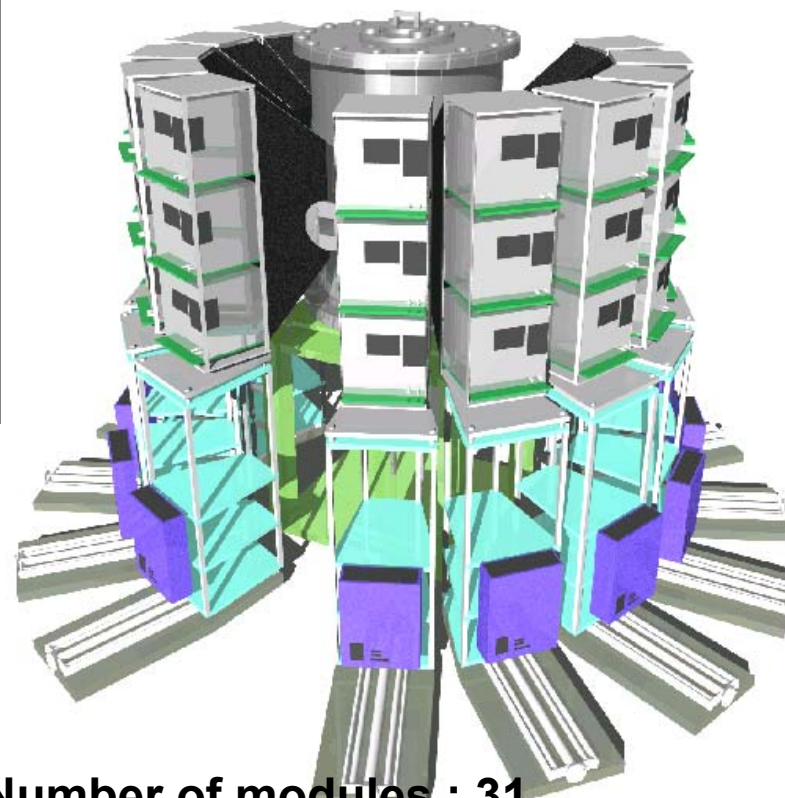
A wide area scintillator detector has been developed using the iBIX detector technology.

## Detector module

Single X-tal diffractometer “SENJU” at BL18



31 modules were fabricated, evaluated and installed.

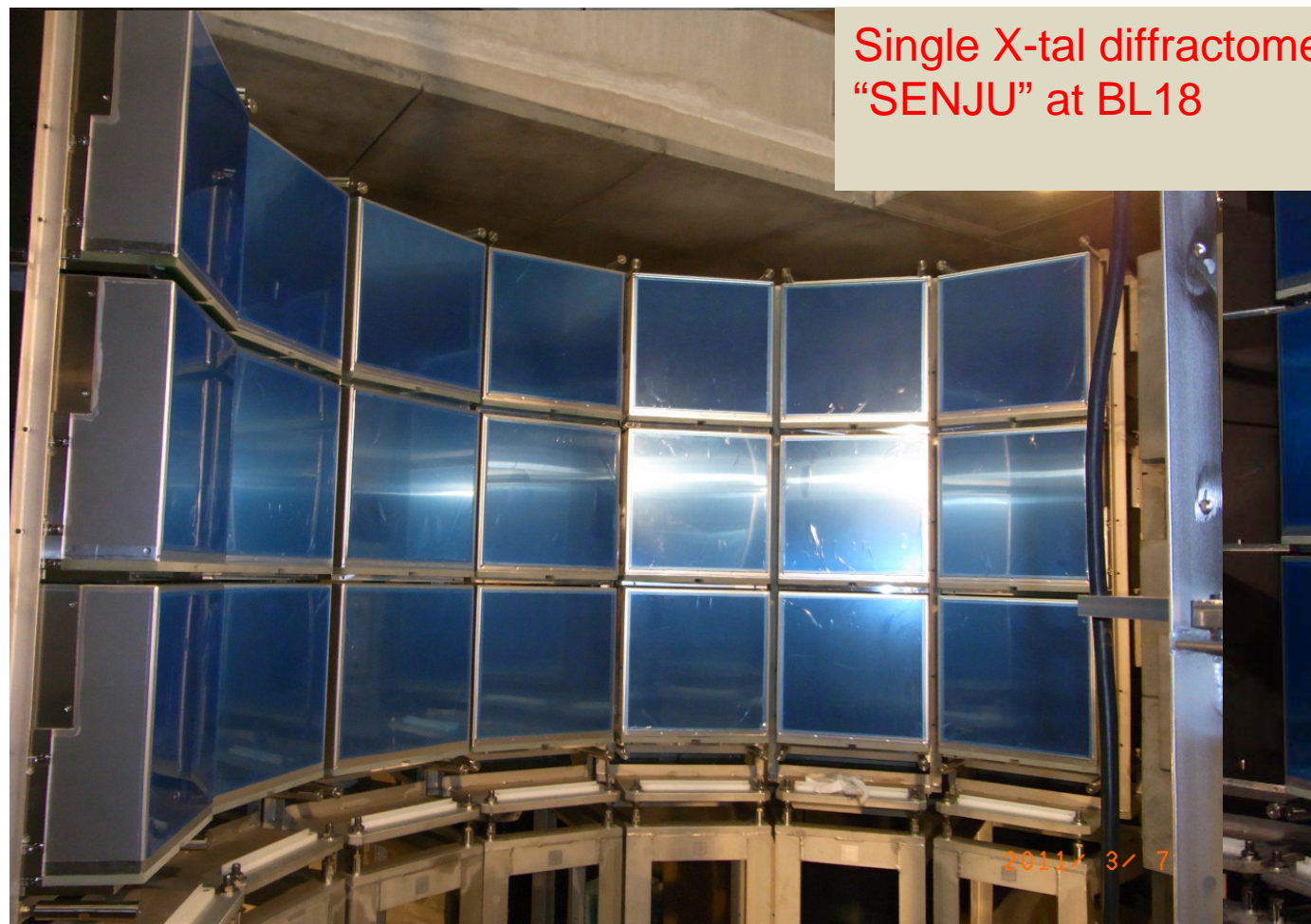


Pixel size	: 4 × 4 mm
Sensitive area	: 256 x 256 mm
Detector efficiency	: 30-40% for 1.8Å
Pulse pair resolution	: < 5 μs
Gamma sensitivity	: ~10 <sup>-6</sup> ( <sup>60</sup> Co)
No. of pixels /detector	: 4096 (64 × 64)
No. of electronics channels	: 128 (64 x 2)
No. of PMTs	: 2

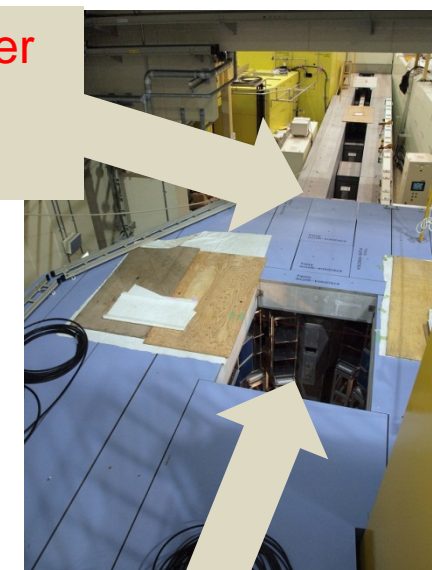
Number of modules : 31  
 Detection area : 2.8 m<sup>2</sup>  
 Pixel size : 4 × 4 mm



# 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  $^3\text{He}$  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

- $^6\text{LiF/ZnS}$  and  $\text{B}_2\text{O}_3/\text{ZnS}$  scintillation detectors with WLS-fibre readout
- Solid  $^{10}\text{B}$  converter in gaseous detectors
- $\text{BF}_3$ -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  $^3\text{He}$ -detectors.

- Efficiency
- Count rate capability
- Cost issues

# Helium-3 Supply - Present Status

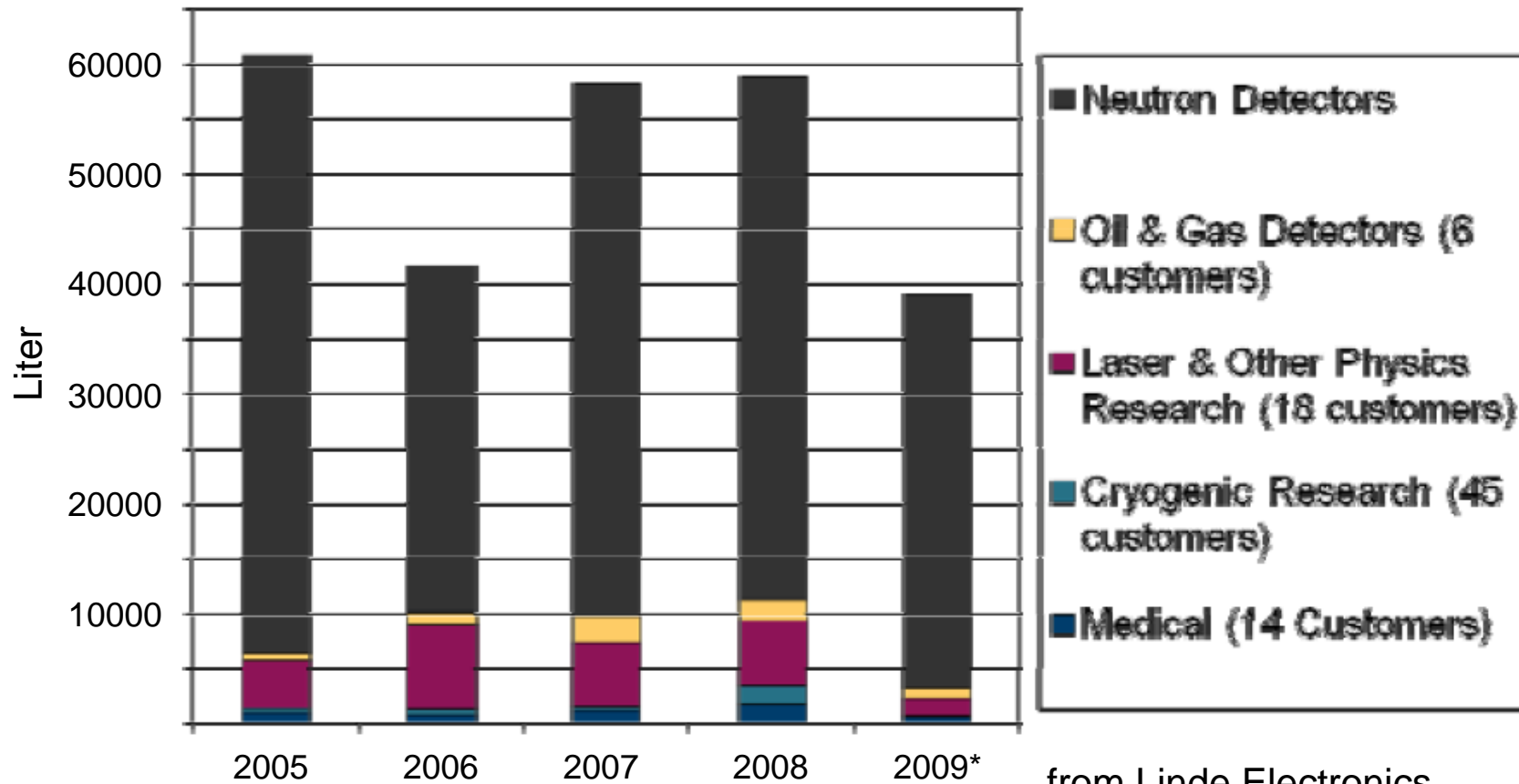
## Interagency Committee set up to steer rationing and allocation of US Helium-3

- 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

## 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 - a brief reminder

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

- Cryogenics / Low temperature physics  $<1^{\circ}\text{K}$
- 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**

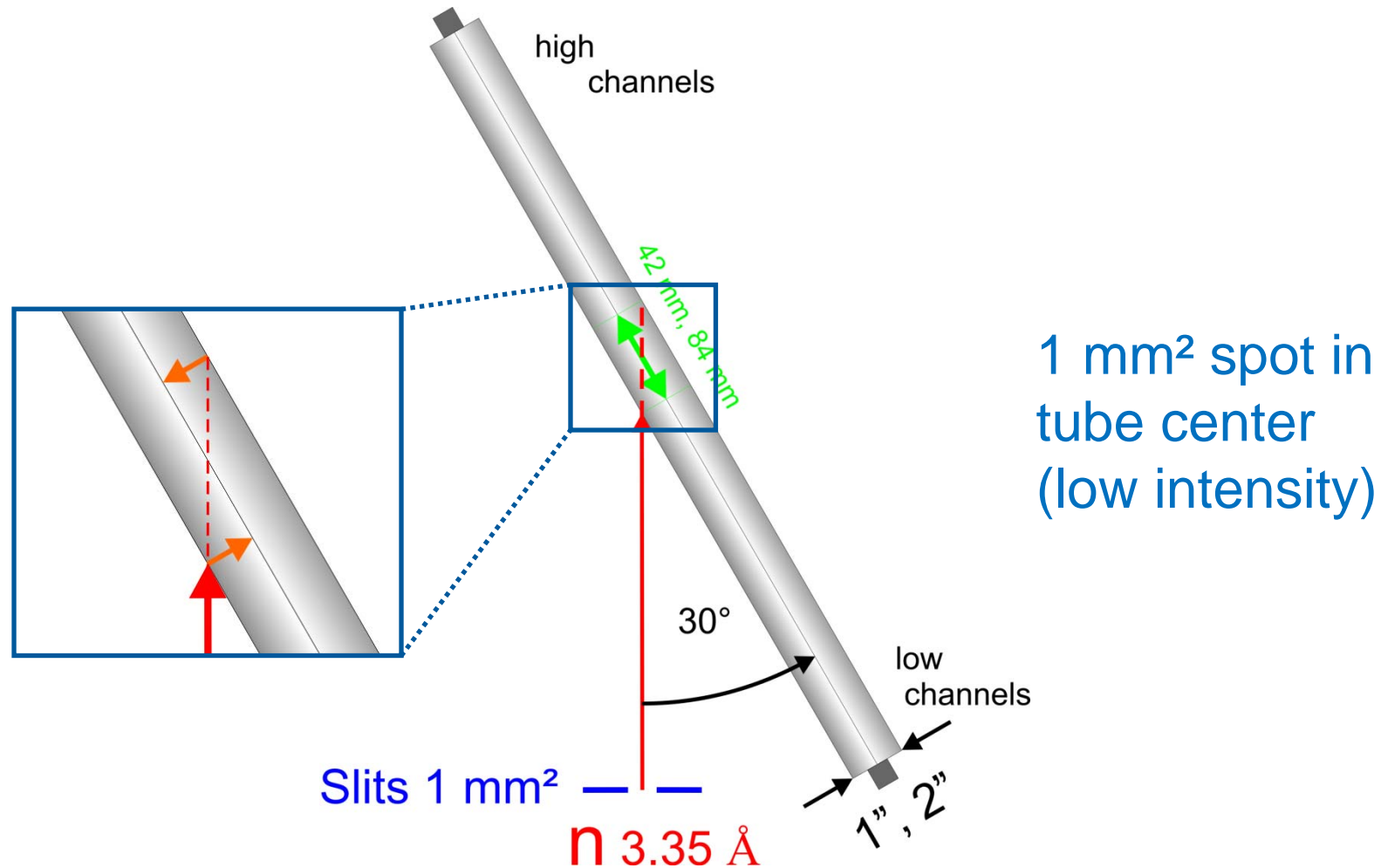
5328b thermal neutron capture x-section + high pressure operation

→ high efficiency, good  $\gamma$  / n separation  $< 10^{-6}$ , inert

mainly used for:

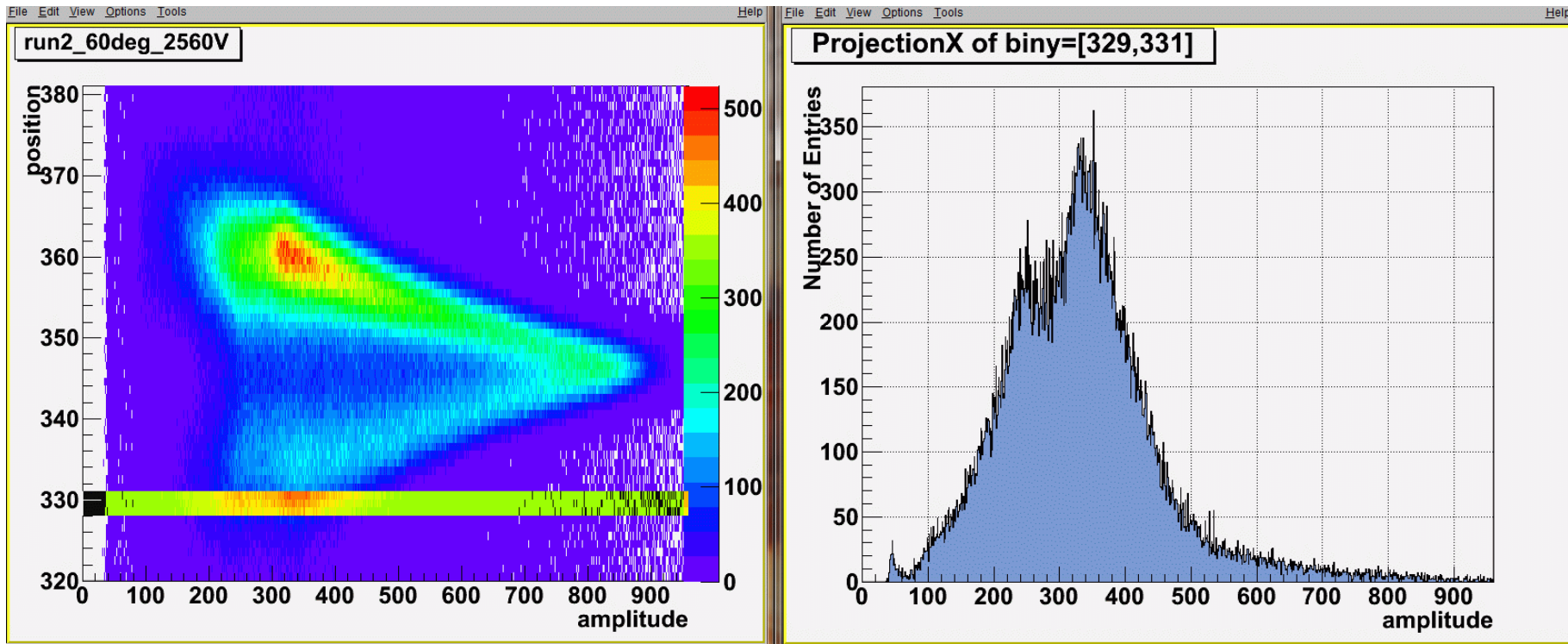
- US homeland security and non-proliferation programmes
- Neutron scattering applications
- Oil well logging and road construction

## Pulse Height vs Amplitude @60° Inclination





# BF<sub>3</sub> Tube (1") – 60° Run Pulse Height Distribution



HV 2560 V,  $\Delta x = 8$  mm fwhm

