# Neutron Detection Alternatives to $^3He$: $BF_3$

**Thomas Wilpert**

**Helmholtz-Zentrum Berlin**

# $^{10}BF_3$ Detectors

- Motivation: NEAT Upgrade
- Brief historical review
- Properties of $^{10}BF_3$ vs. $^{3}He$
- Results of measurement with IN5 prototype
- Results of measurements with LPSDs
- Concept for detectors for time-of-flight spectrometer
- Safety measures
- Results of measurements with 1st HZB Prototype
- Conclusion

# Time-of-Flight Spectrometer

Requirements on detectors for modern ToF Spectrometer

- Large angular coverage (horizontal and vertical)
  - Several dozen $m^2$ active area
- 2D position information
- High efficiency
- Good ToF resolution
  - Depth ≤1”
  - Flight path ≥ 3 m
- Operation in vacuum

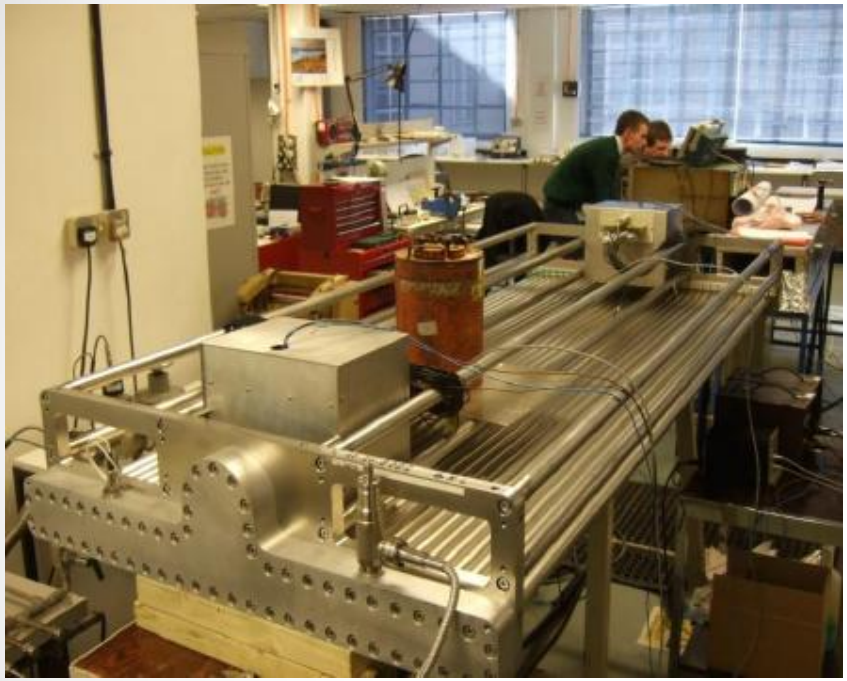
# Time-of-Flight Spectrometer

LET at ISIS

**4 m long $^3$He tubes from GE-RS**

- Longest tubes ever built
- Resistive anode wire
- 10 bar $^3$He pressure
- 7800 liter $^3$He (value today ~19 M€)

# Time-of-Flight Spectrometer



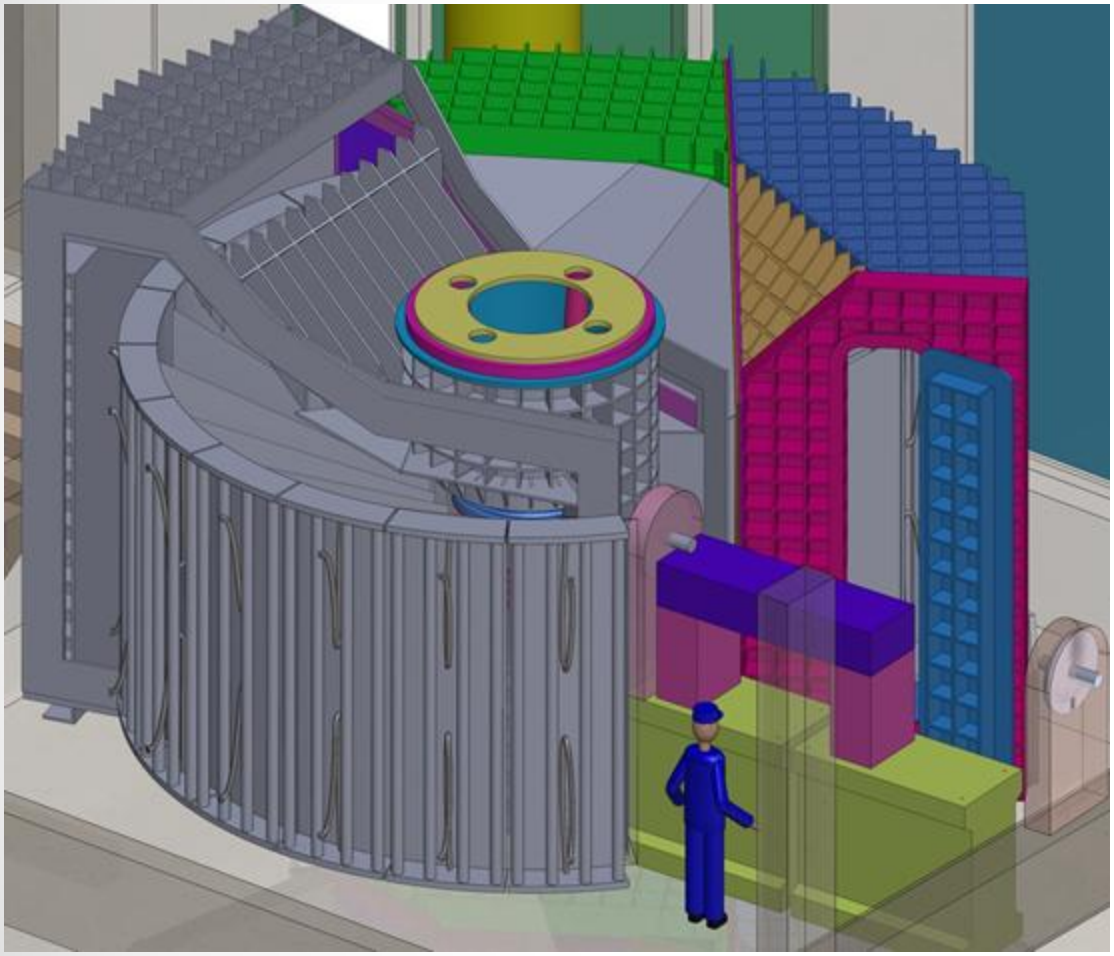
IN5 at ILL

**“Multitube” design developed at ILL**

- Tube ends welded in common flange
- All tubes share the same gas volume
- Stainless steel resistive anode wire

- 3 m long 1” LPSDs, 4.5bar $^3$He,
- ~3000 liter $^3$He in total

# Time-of-Flight Spectrometer



## NEAT-upgrade project

- **Same performance as IN5 (less n-flux → larger solid angle)**
- **Pixel resolution of 2.5 x 2.5 cm²**
- **TOF resolution <30 μs (for cold neutrons)**
- **90% efficiency @ 5Å**
- **Radius of detection plane 3 m**
- **In-plane acceptance angle 280°**
- **Active height 3 m**
- **Active detector area ~40 m²**
- **17 Modules, each 32 1" tubes**
- **544 tubes in total**
- **Replace equivalent of ~3300 liter $^3$He**

**Ready in 2015!**

# Why $BF_3$?

- Scintillation detectors?
  - price out of scope for NEAT
  - no tradition/knowledge at HZB

- $^{10}B$ multi layer?
  - time for coating 6.3 m²: 2 weeks
  - 40 m² required (20 layers!): 126 weeks (~2.5 years)
  - no experience in real experiments
  - stability, ageing?
  - costs? 2012: 1500 EUR/m² only coating

- LPSDs filled with $BF_3$
  - Same detector technology as for $^3He$ tubes (higher HV!)
  - Adopted detector design to compensate less efficient tubes
  - Price for $^{10}BF_3$ (neutron detector grade) ~same as for $^3He$ before shortage
  - $BF_3$ is widely used in chemical industry (catalyst, base mat. lubricants, fragrances, … )

# Brief Historical Review

| | |
|---|---|
| 1939: | First use of $BF_3$ by Korff and Danforth (slow neutrons in cosmic radiation) |
| 1948: | $^{10}BF_3$ detector in powder diffraction by E.O. Wollan and C.G. Shull at ORNL |
| 1953: | $^{10}BF_3$ commercial product by 20th Century Electronics |
| 1963: | Very large tube built by Fowler (14.5 cm dia., 183 cm long) |
| 1970s-1980s: | „Banana“ detectors for diffraction experiments, SANS detectors D11(ILL), V4 (HZB) by LETI/France |
| 1985 - 2005: | Replacement by $^{3}He$-filled tubes, MWPCs |
| 2009: | Reconsideration of $BF_3$ for time-of-flight spectrometer NEAT/HZB |
| >1950 | used as startup detector in nuclear power plants (good n/$\gamma$ discrimination) |

# Brief Historical Review

~1980s: $BF_3$ filling of banana detector by LETI
(disposed 2008)

# Properties $^{10}BF_3$ vs. $^{3}He$

| Isotope | Reaction | 1.81 Å Cross Section [b] | Absorb. Length [bar·cm] | Product Energies [keV] | Product Range [cm] | Chemical |
|---|---|---|---|---|---|---|
| $^{3}He$ | $^{3}He(n,p)t$ | 5333 | 7.59 | p 573<br>t 191 | $R_p = 0.43$ ($CF_4$) | nobel gas (inert) |
| $^{10}BF_3$ | $^{10}B(n,\alpha)^{7}Li$ | 3836 | 9.82 | α 1472<br>$^{7}Li$ 840<br>93% | $R_\alpha = 0.42$ | toxic reacts with water (HF) |

## $^{3}He$

**High cross section**
**p ≤15 bar possible**
**Noble gas**
**Easy to use**
**Very pure**
**LPSD standard**
**But: stopping gas req.**

## Toxicity

| Compound | MAC* Value |
|---|---|
| $H_2Se$ | $6.0 \times 10^{-9}$ |
| $BF_3$ | $3.5 \times 10^{-7}$ |
| $H_2S$ | $5.0 \times 10^{-6}$ |

*MAC maximum allowed concentration

## $^{10}BF_3$

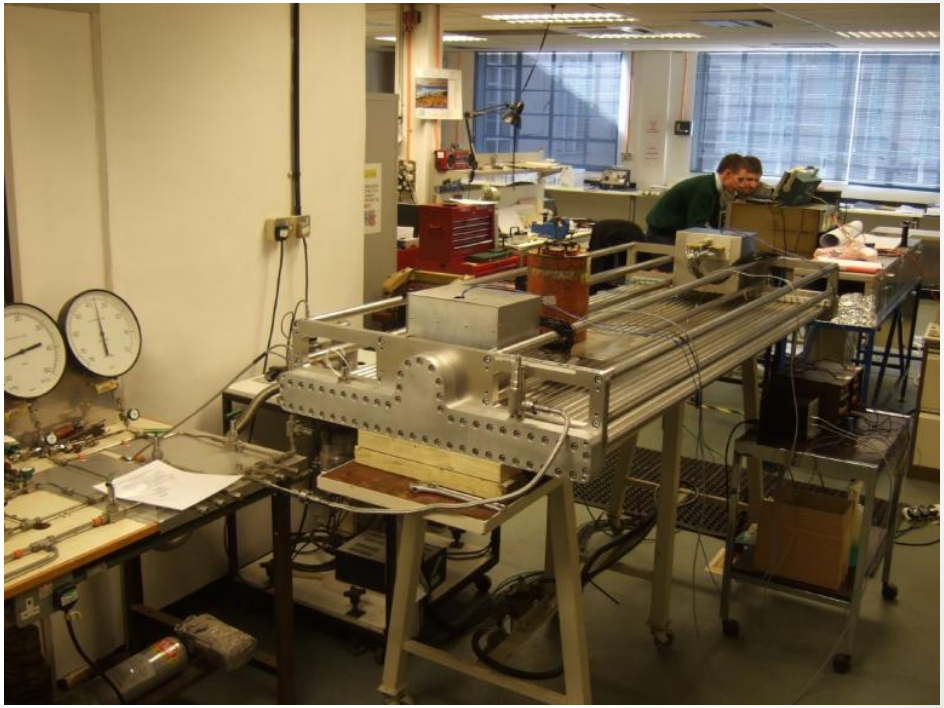
71.9% cross section of $^{3}He$
p ≤ 2 bar
Highly reactive gas: HF
Toxic, irritant, pungent smell
Impurities!
LPSD (to be proven)
Transportation (some effort!)

# $BF_3$ - Tests with IN5 Prototype

Quick test of IN5 module (2 m, flat) at Centronic, Croydon/UK

Observations

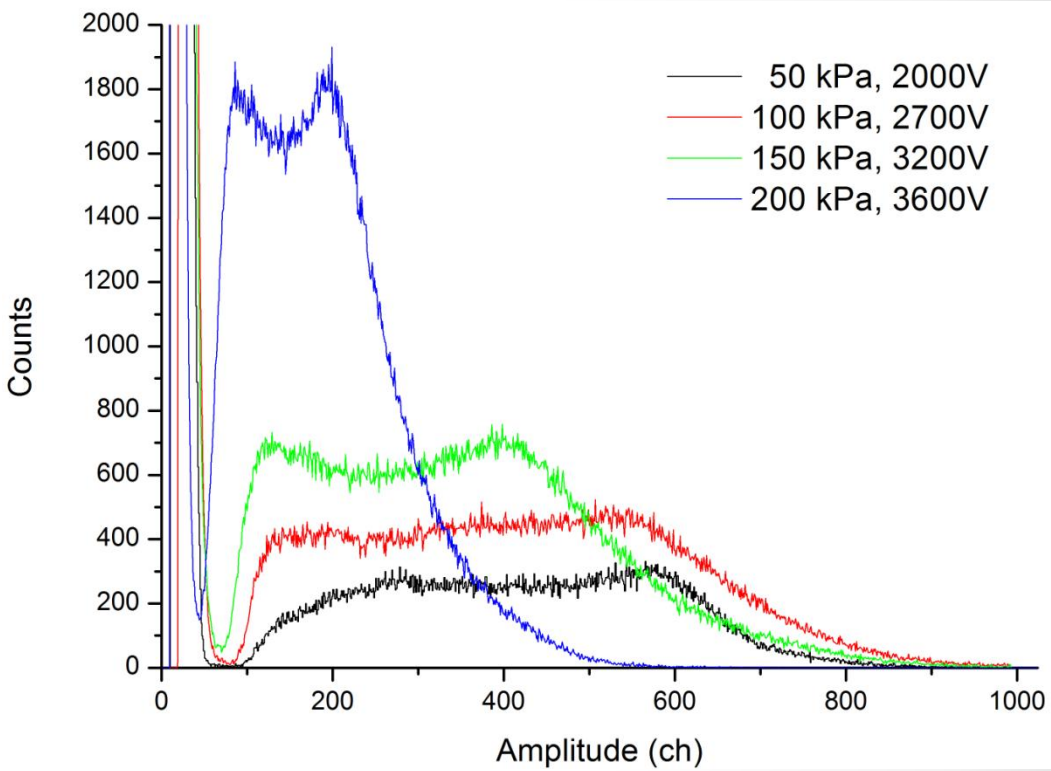
- Pulse height spectrum deteriorates with increasing pressure (up to 2 bar)
- High voltage had to be increased beyond electronics capability of 3 kV



Thanks to B. Guerard (ILL)

# $BF_3$ - Tests with IN5 Prototype

- 2 m long IN5 module (32 tubes) filled with $^{10}BF_3$ at company Centronic/UK
- 2 bar max. (99% $^{10}BF_3$), 3600 V max. tested with ILL electronics
- First tests with $^{241}$Am-Be source made on March 2-3, 2010
- 2D operation was demonstrated



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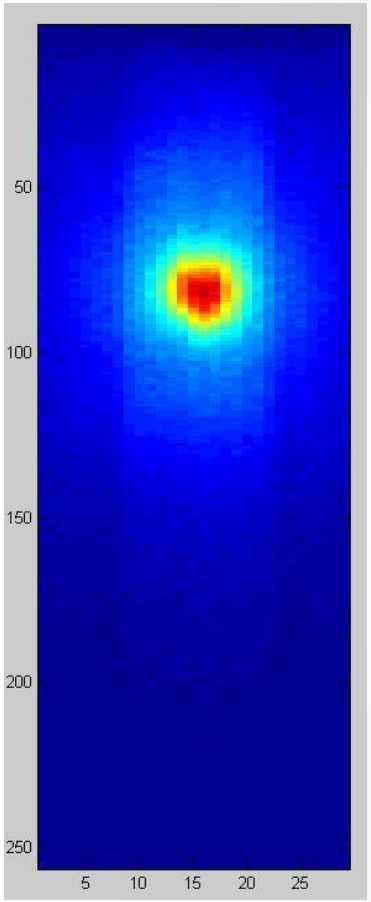
Results are encouraging!




2 bar, 3600 V

# $^{3}He$-$BF_3$ – Comparative Tests

| Company | Isotope | Pressure [bar] | Diameter [cm] | Length [cm] | Resist. [kΩ] | Year |
|---|---|---|---|---|---|---|
| Centronic | $^{10}BF_3$ (99%) | 1.87 | 2.54 | 100 | 6.0 | 2009 |
| | | | | | | |
| 20th Century Electronic | $^{10}BF_3$ | 0.97 | 5.08 | 56 | 1.6 | 1974 - 1977 |
| | | | | | | *) |
| Dextray | $^{3}He$ | 4.3 | 2.54 | 45 | 3.6 | 2004 |
| | | | | | | |

At first, characterizing $BF_3$ tube

*) Thanks to R. Kampmann, H. Eckerlebe (HZG)

# BF3 Tube - Performance

- Shaping times
- Position resolution dep. on HV
- Position resolution along tube
- Efficiency along tube
- Efficiency across tube
- Gamma sensitivity (tricky, omitted here)
- Pulse height vs amplitude @60  inclination

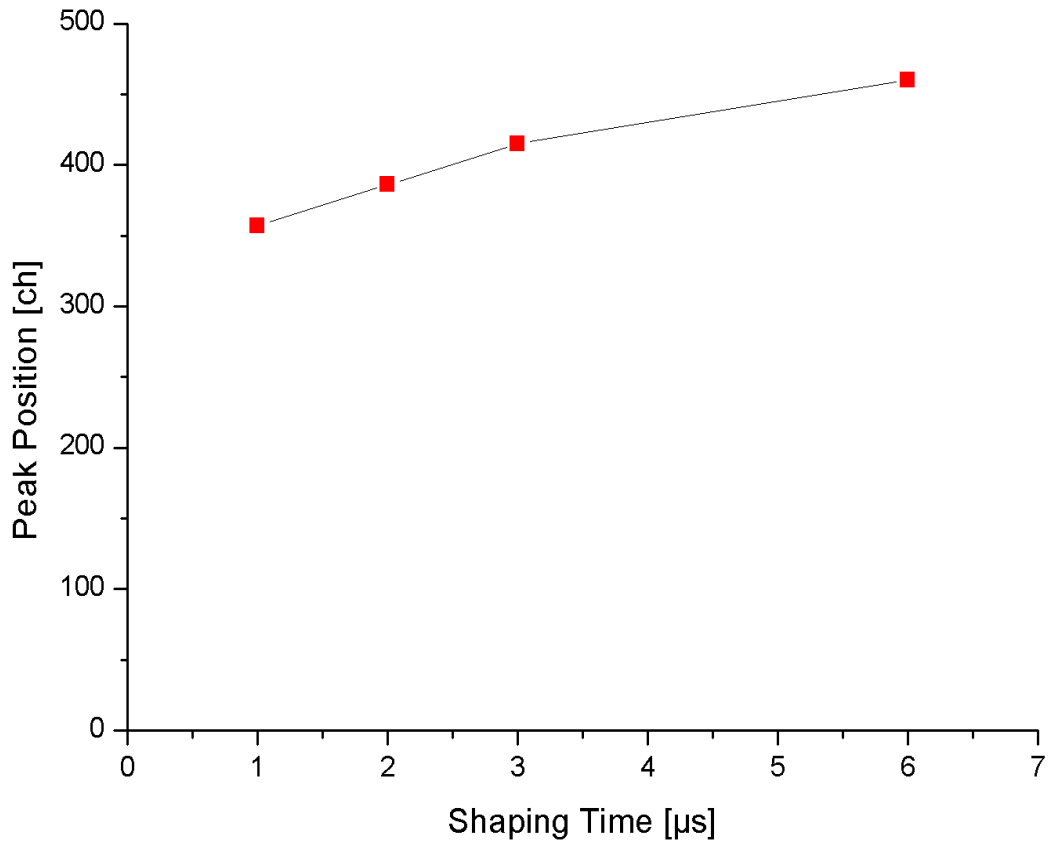
Experiments:

- TREFF at FRM II (λ = 4.74 Å)
- V17 at HZB (λ = 3.35 Å)

Mesytec electronics: MPSD-8+, MCPD-8 *)

*) www.mesytec.com

# $BF_3$ – Shaping Times




Operation Parameter:

1 mm spot

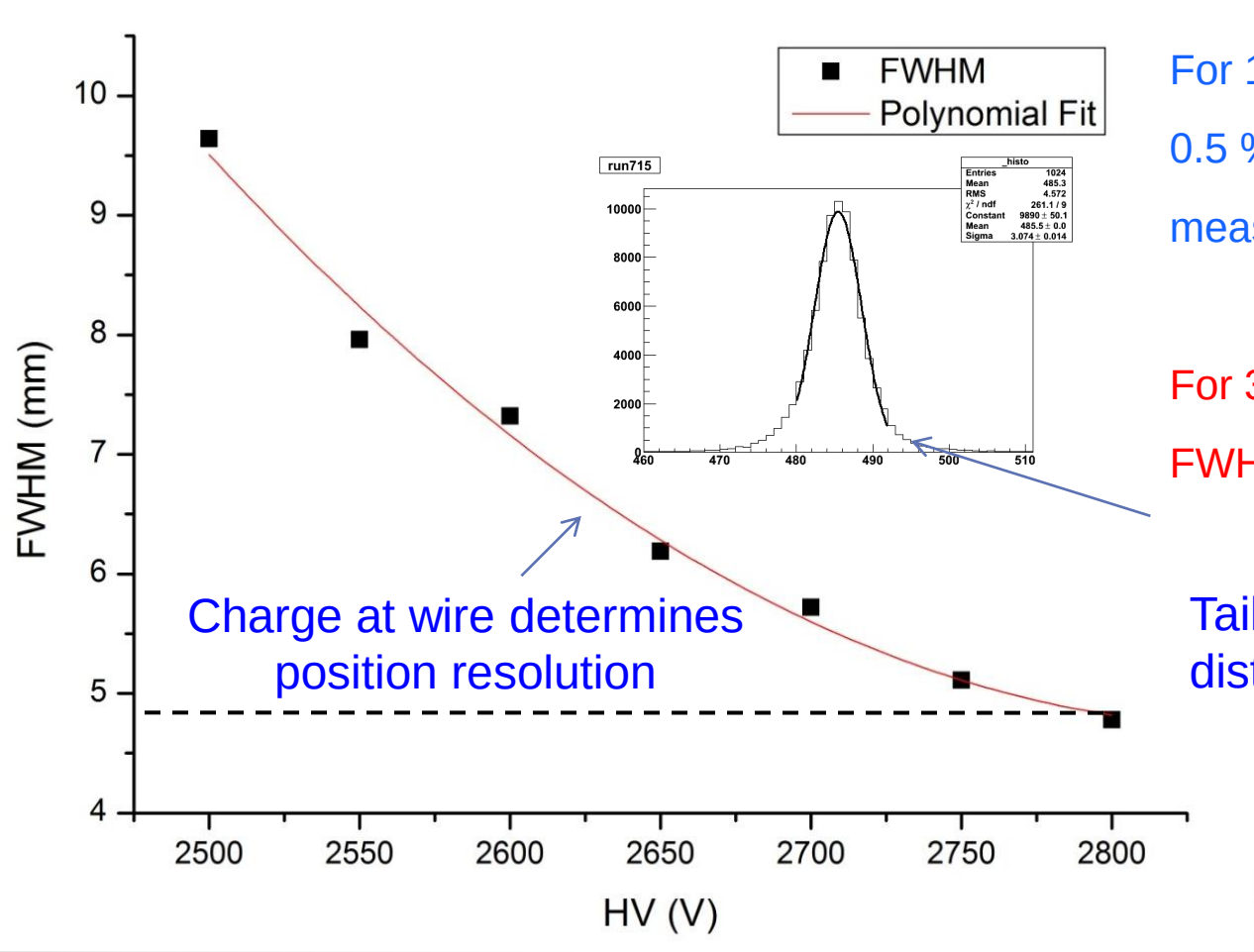
HV: 2750 V

preamp: Canberra 2006

shap. amp: Silena 7614

Standard electronic with
1 µs shaping can be used

# $BF_3$ – Position Resolution




For 1 m tube

0.5 % position resolution

measured

For 3 m tube

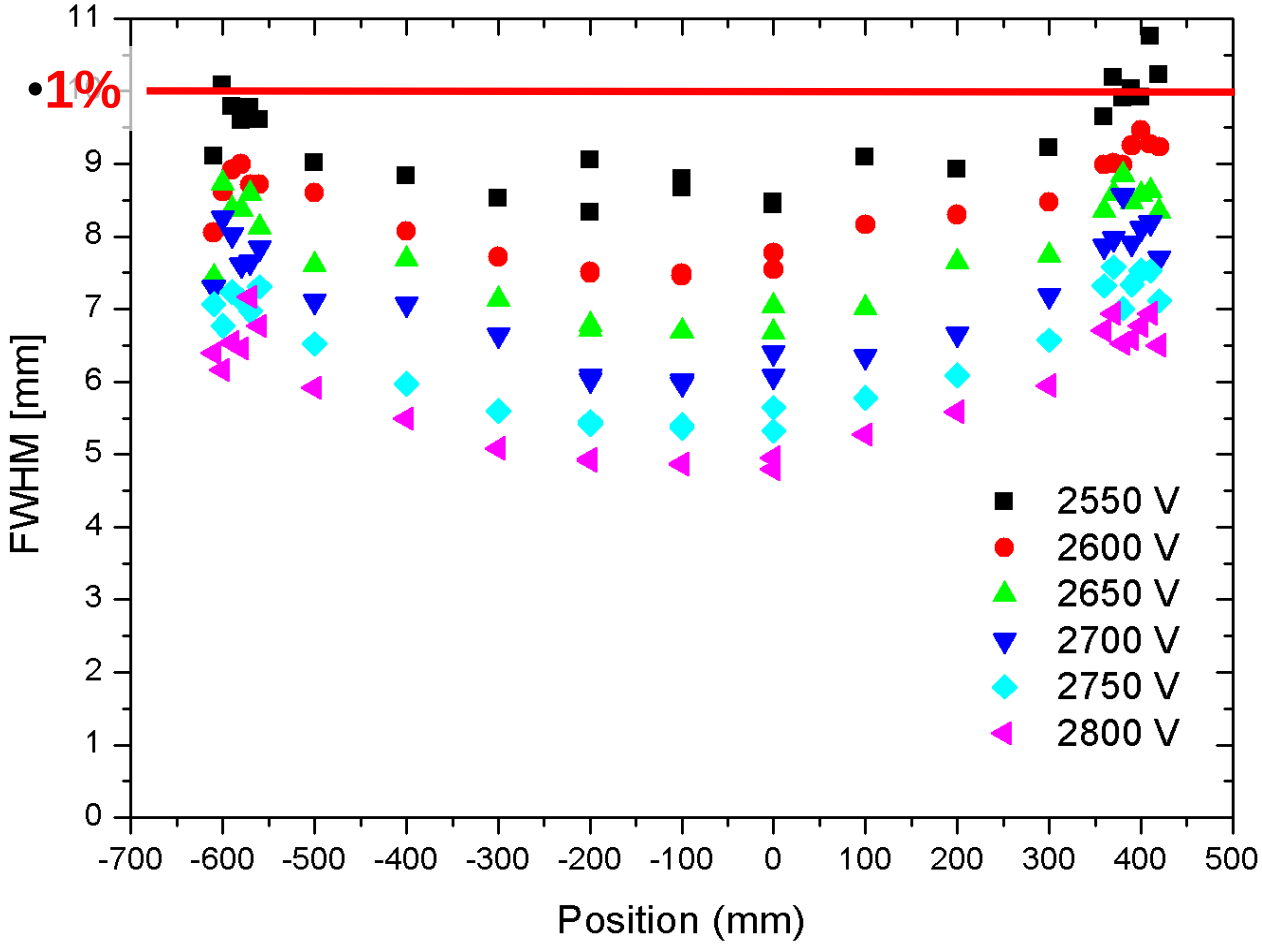
FWHM ~20 mm possible

Charge at wire determines position resolution

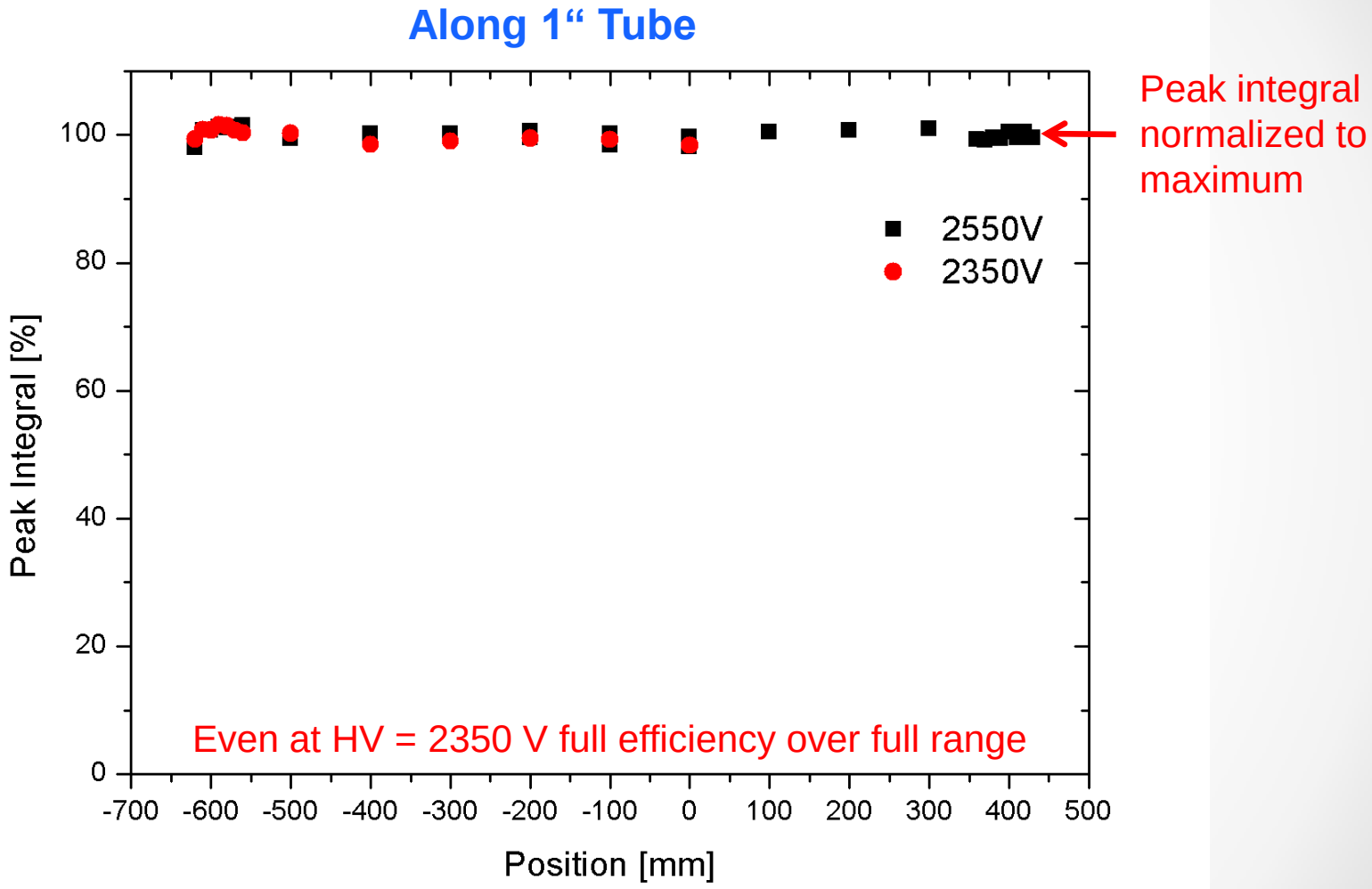
Tails due to amplitude distribution

# $BF_3$ – Position Resolution

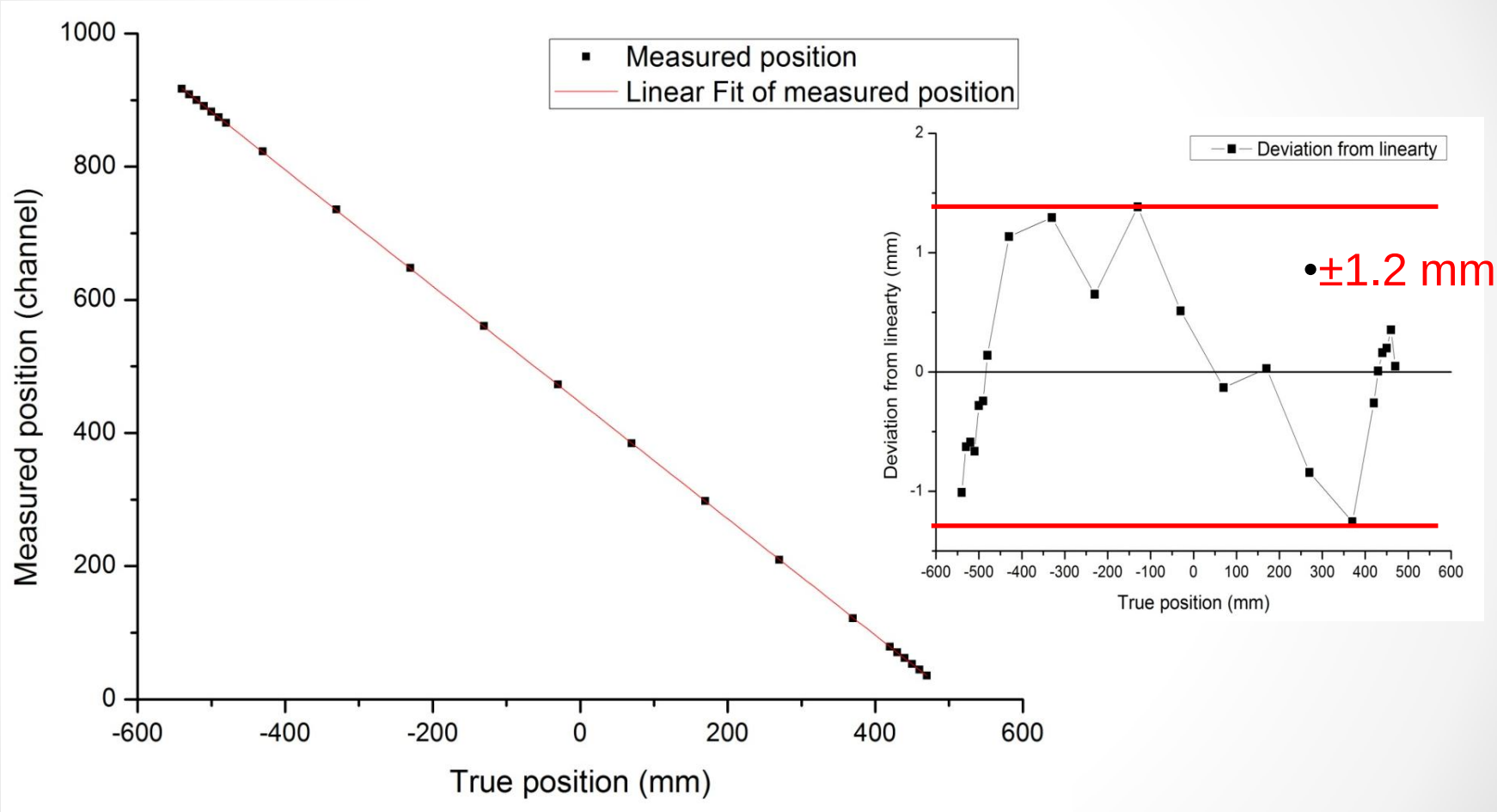
**Along 1“ Tube**



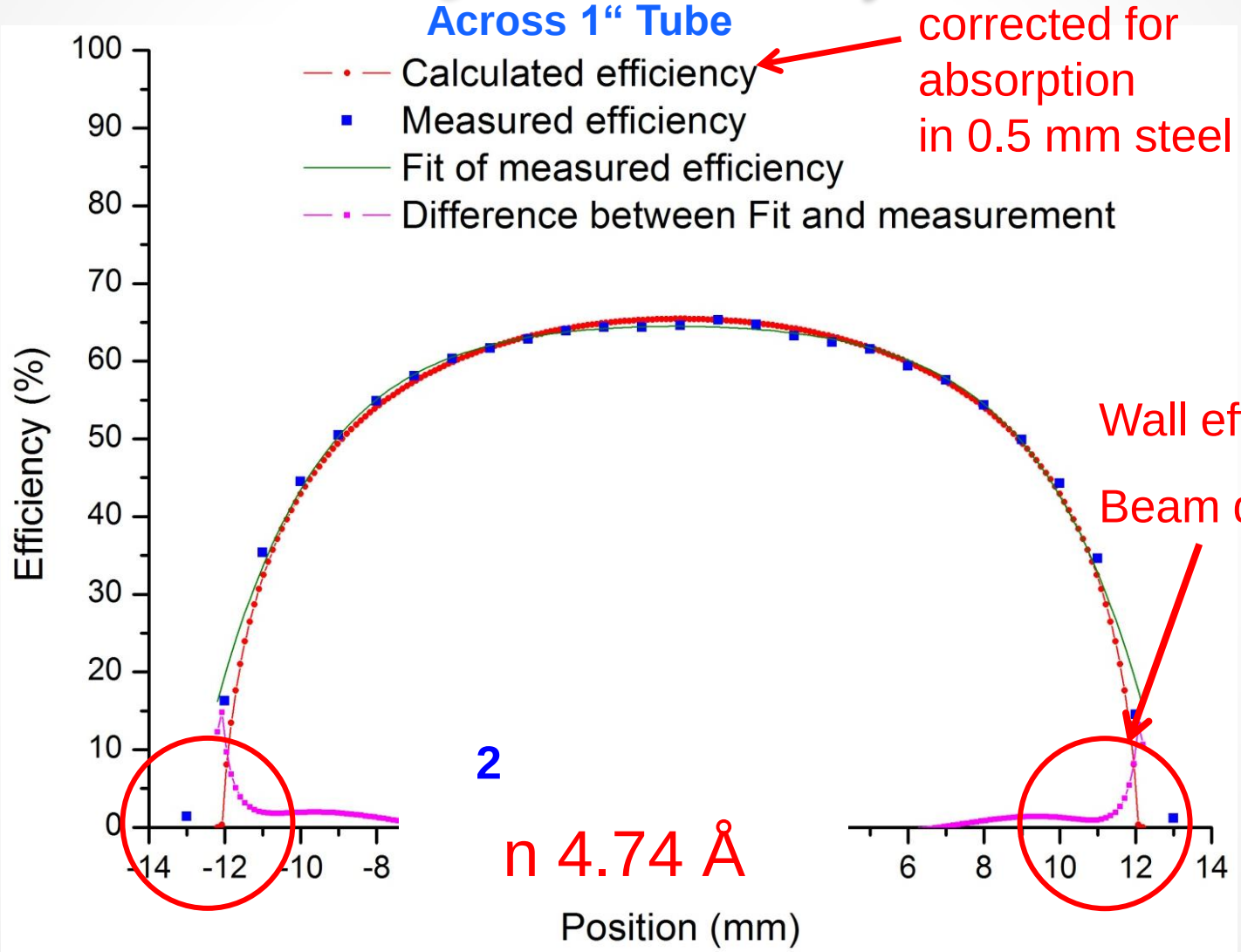
# $BF_3$ – (rel.) Efficiency



# $BF_3$ – Linearity



# $BF_3$ – Effifiency

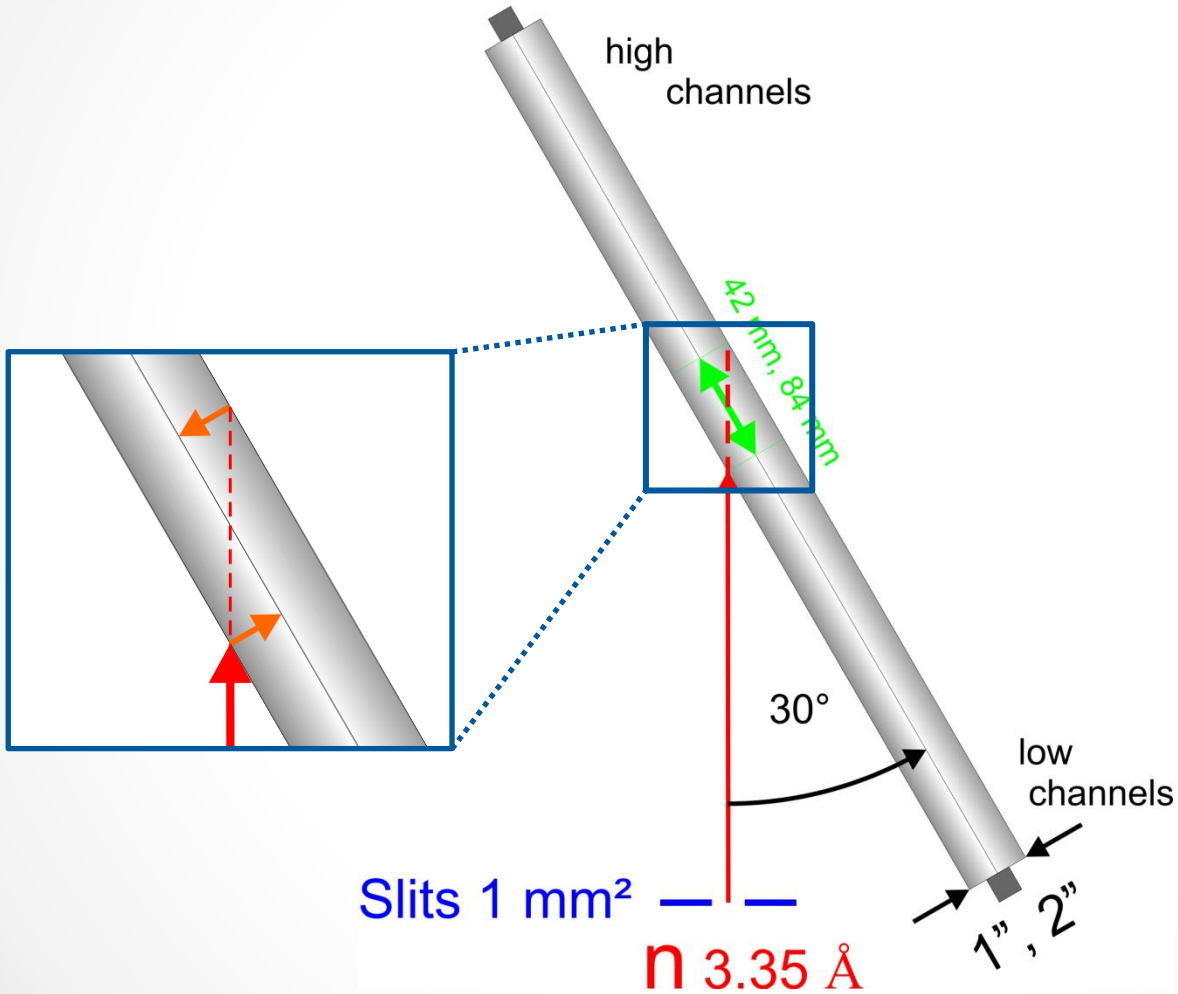



corrected for absorption in 0.5 mm steel

Wall effect +

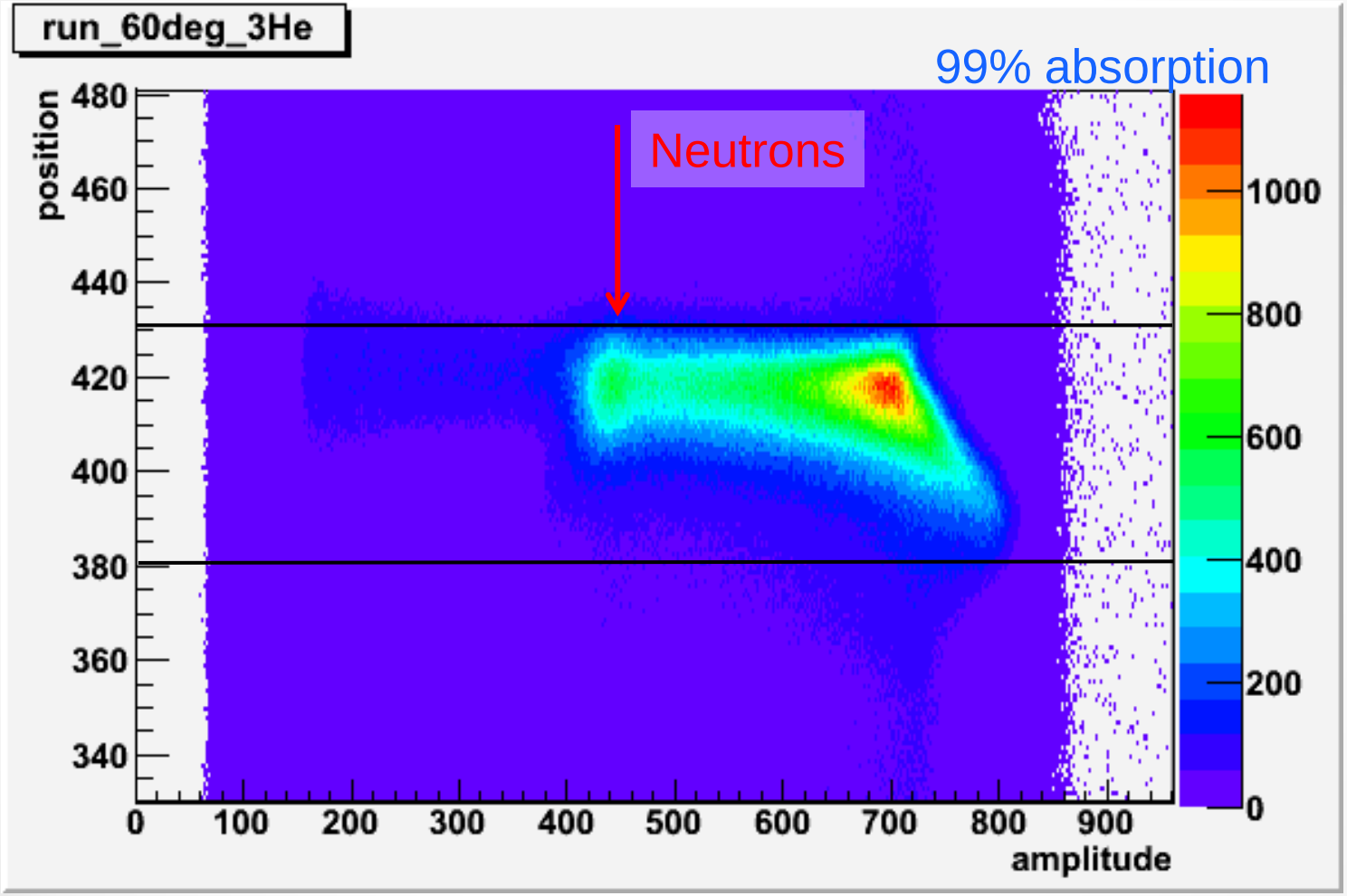
Beam divergence

# $BF_3$ – Gas Amplification




1 mm² spot in tube center (low intensity)

# $^3$He – Reference Measurement



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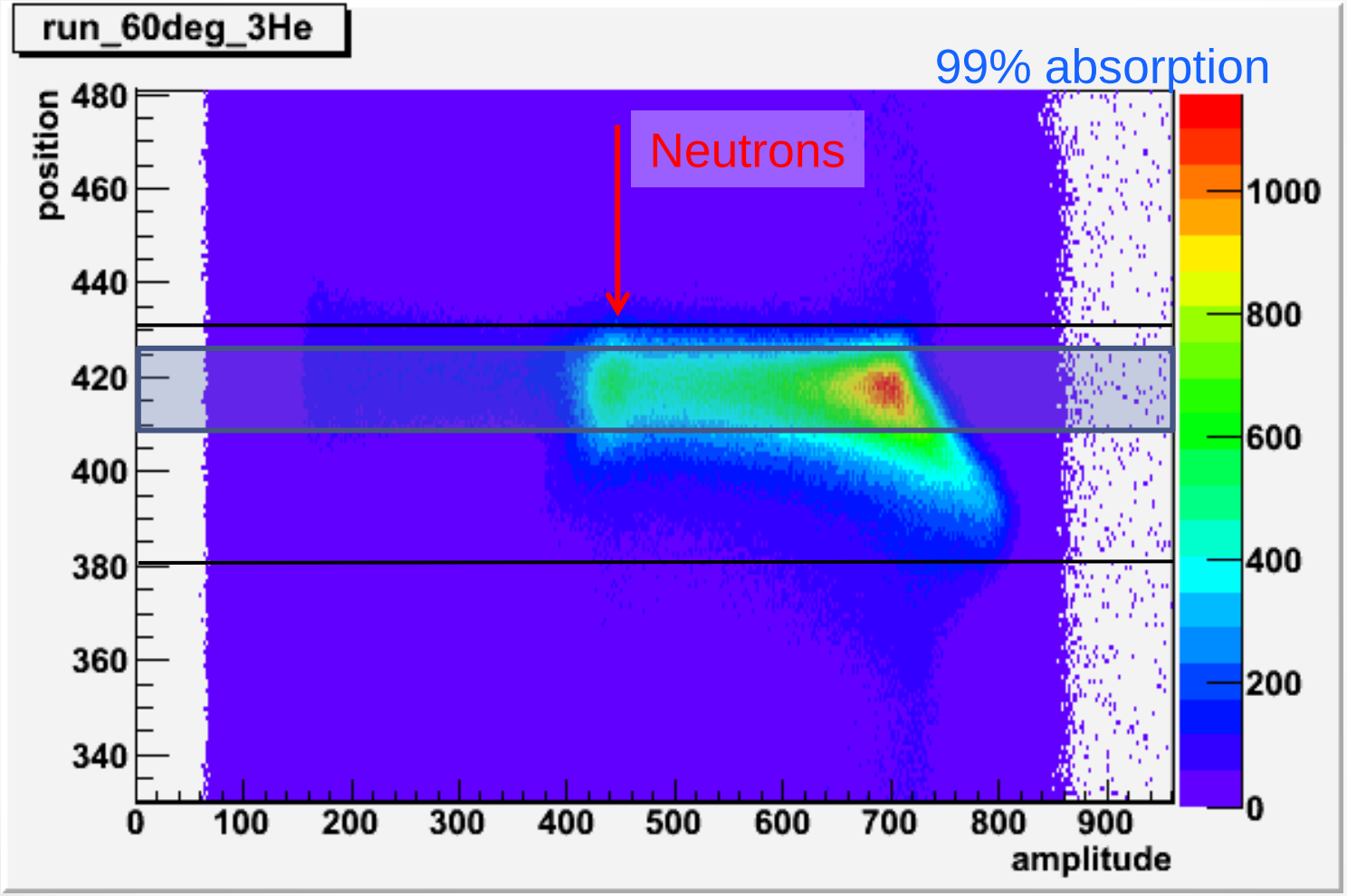

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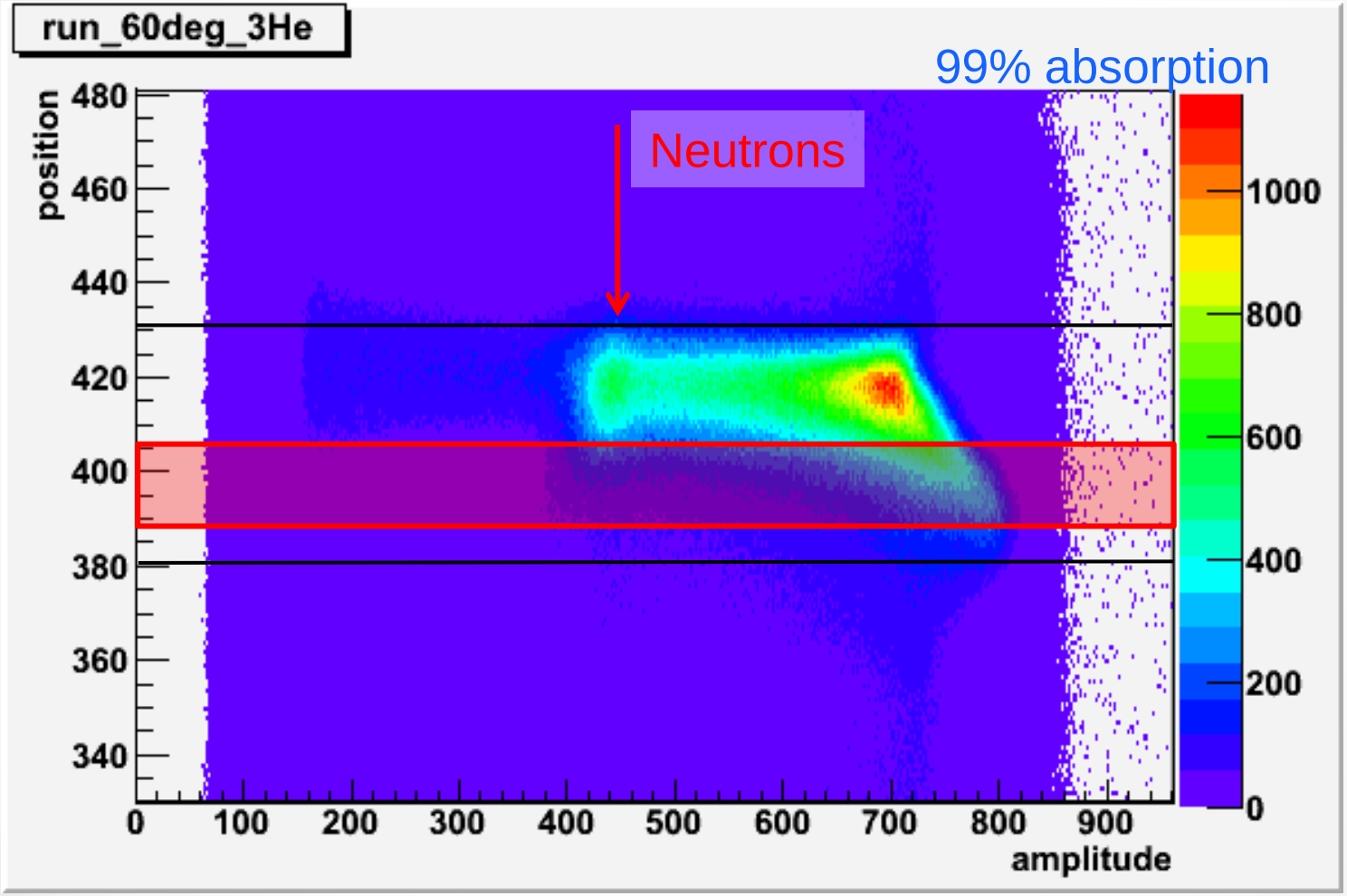

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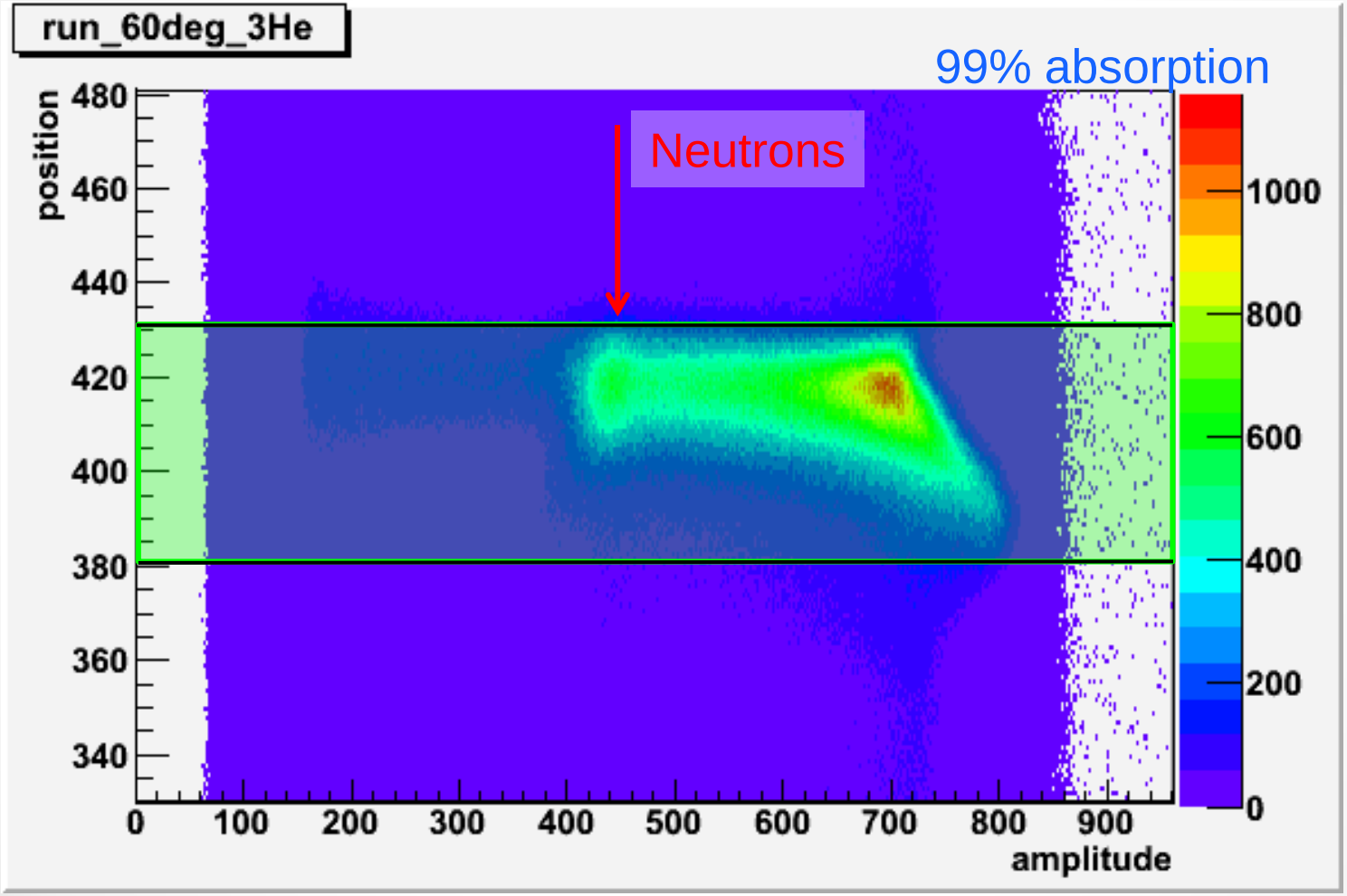
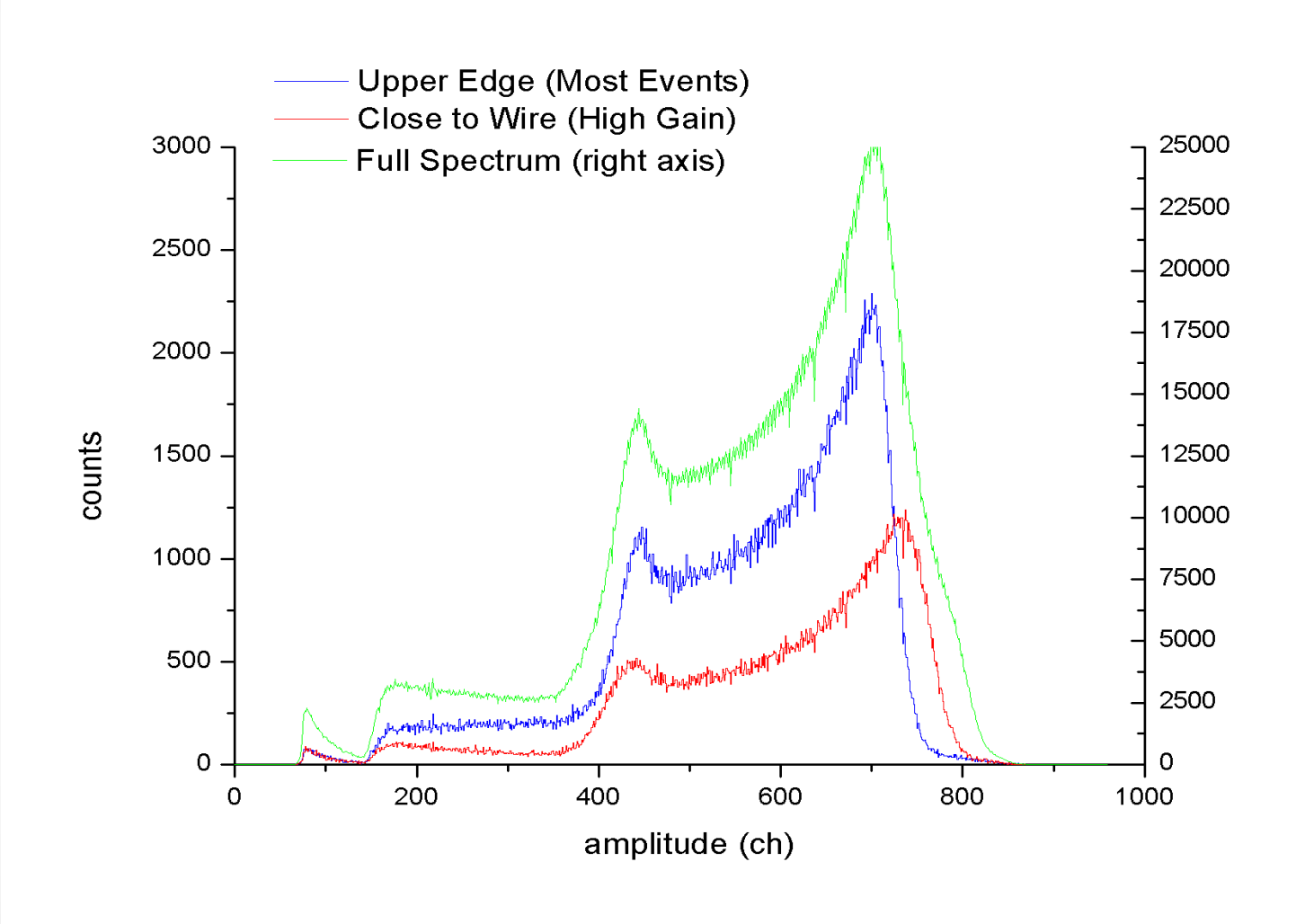
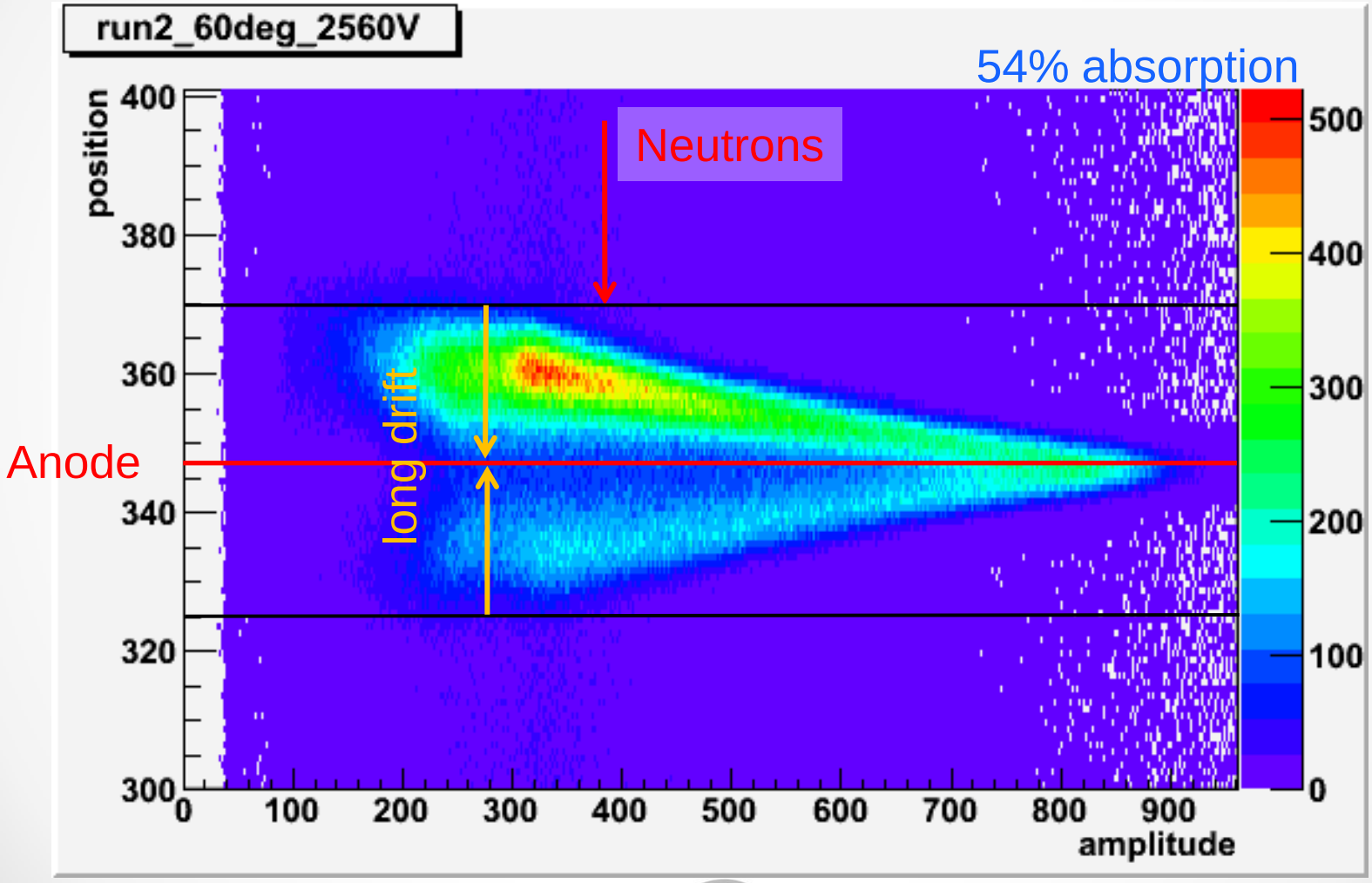


# $^3$He – Reference Pulse Height



# $BF_3$ – Pulse Height (1”)



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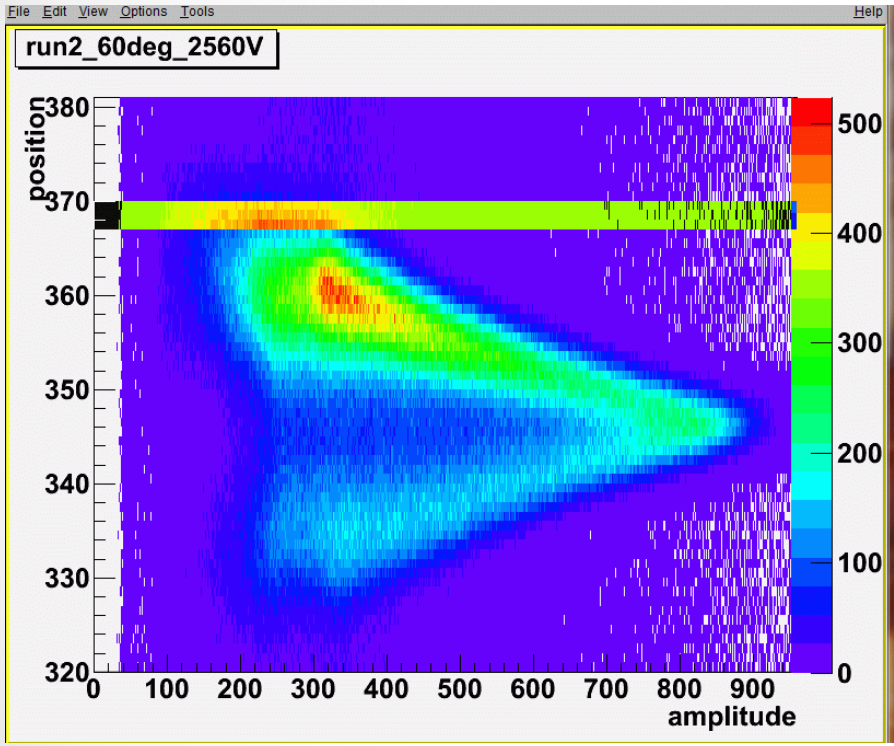


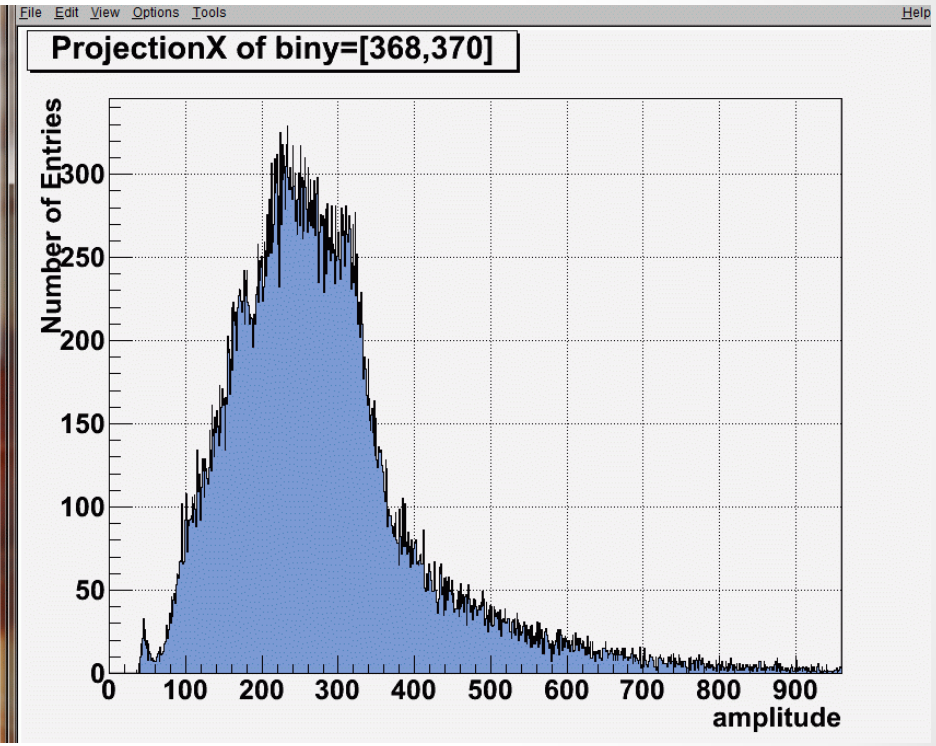


HV 2560 V, $\Delta x$ = 8 mm fwhm

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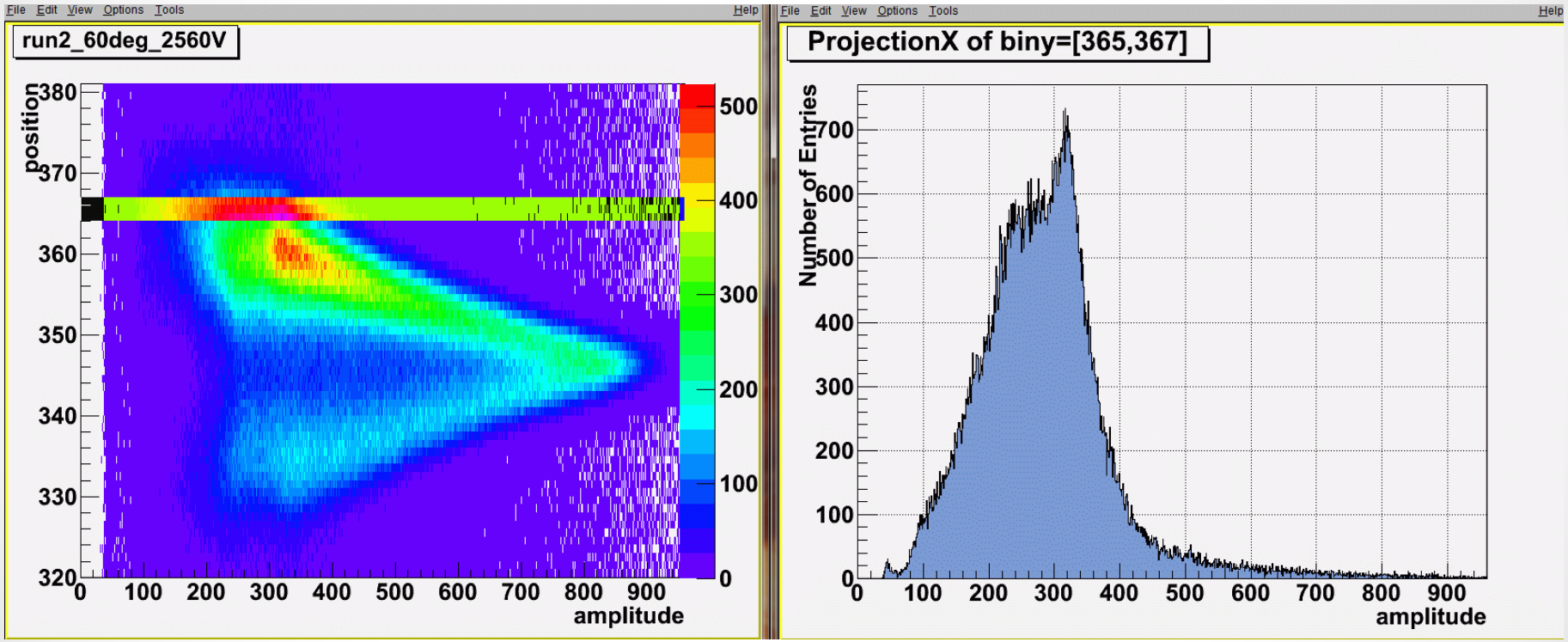


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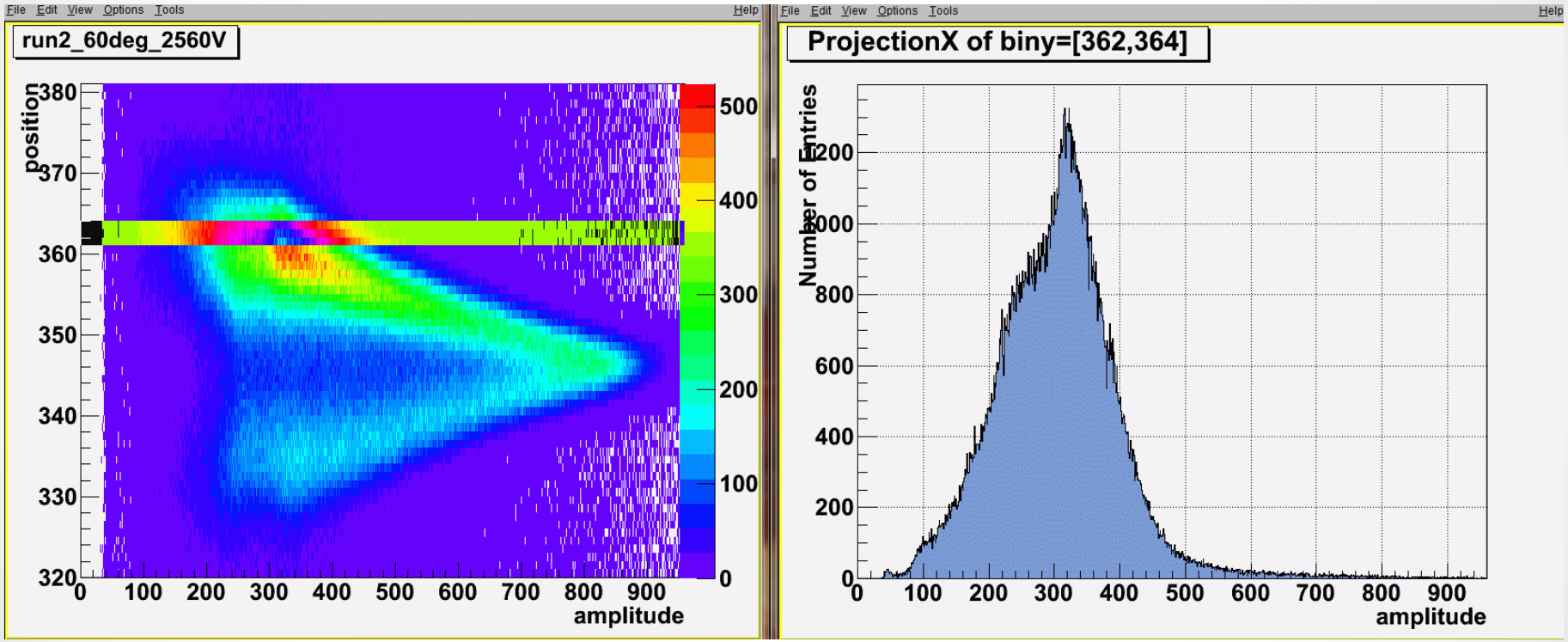


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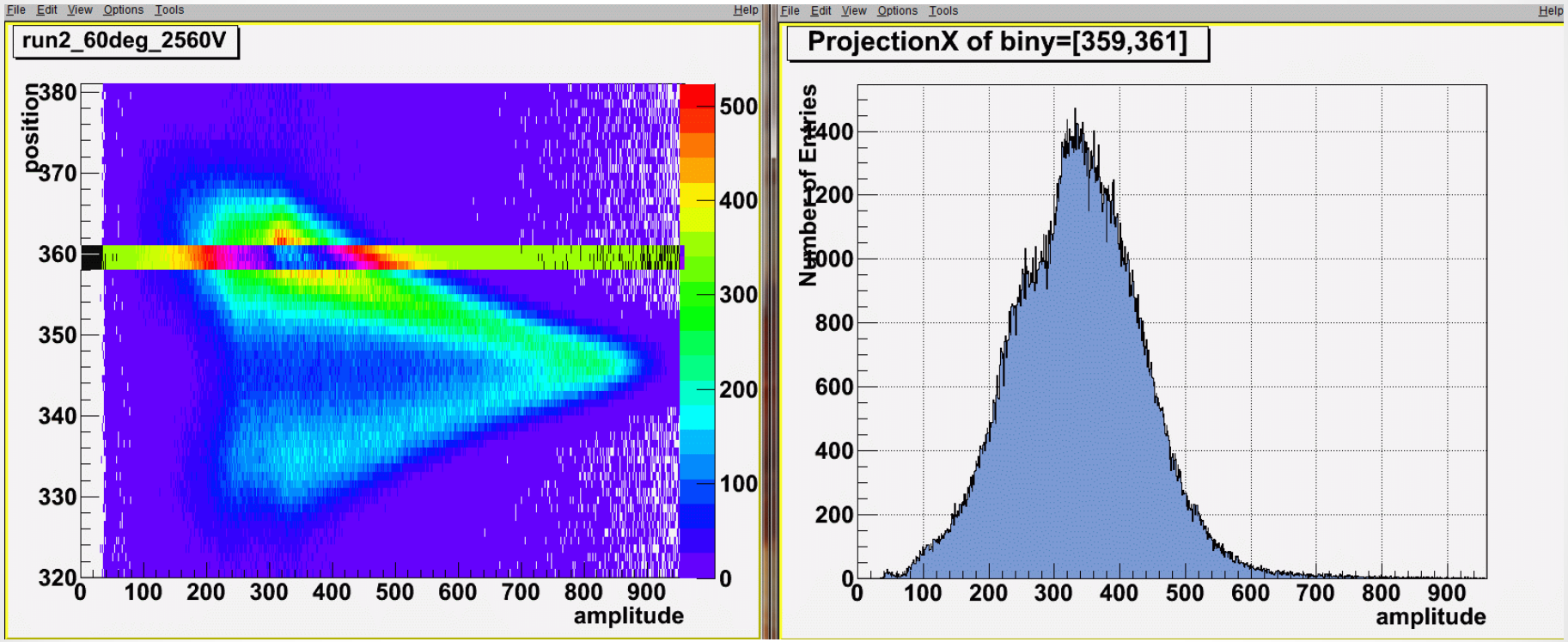


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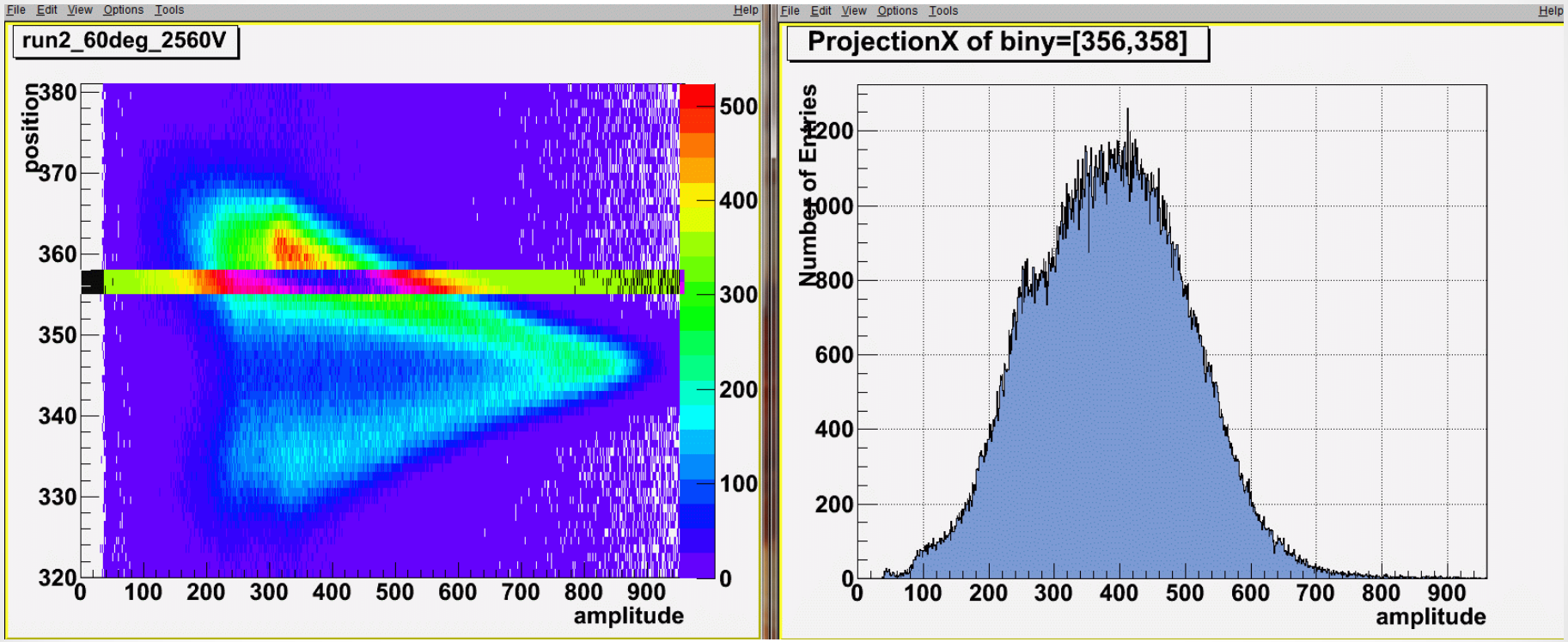


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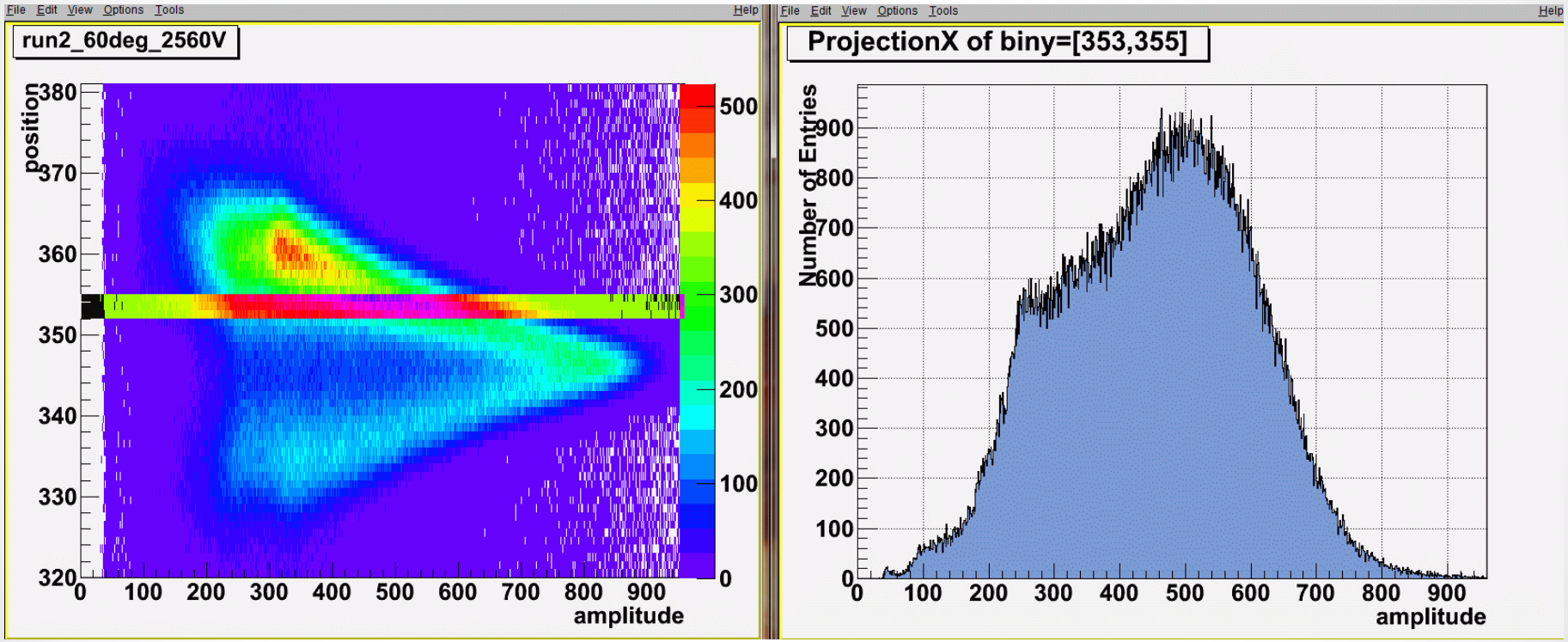


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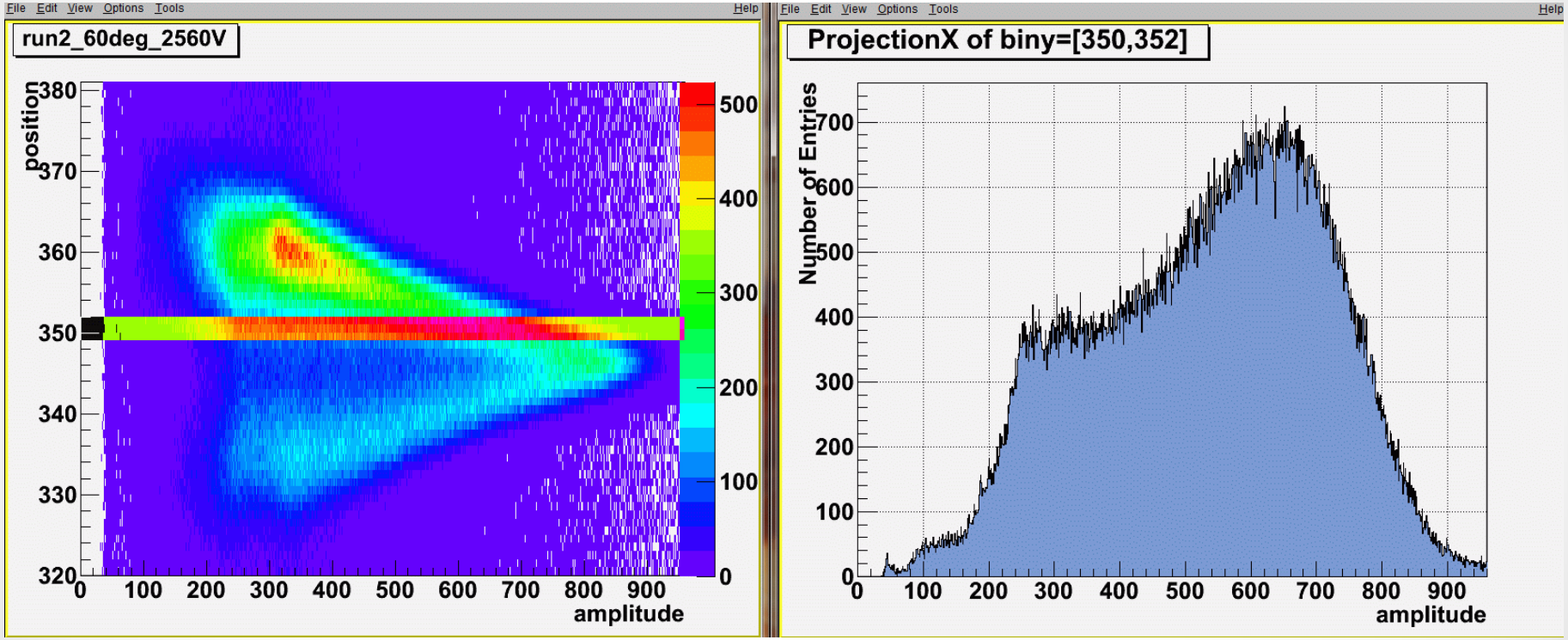


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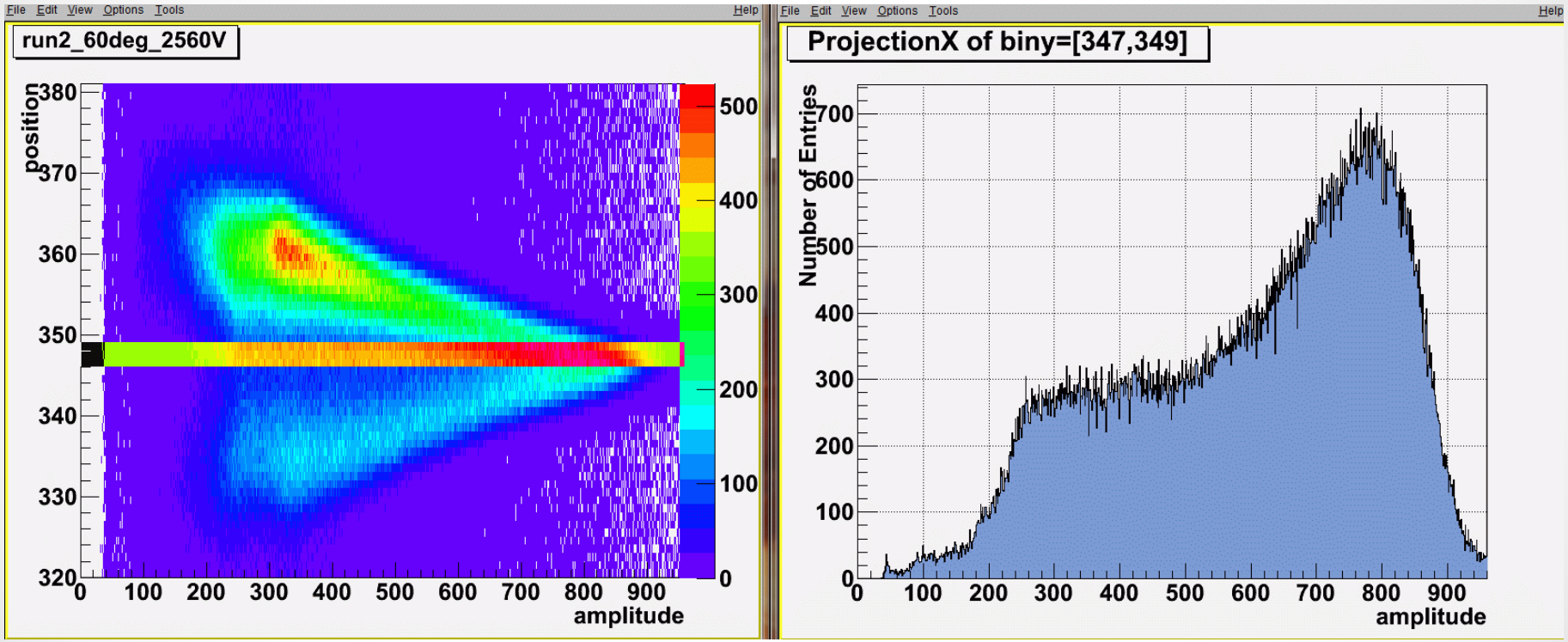


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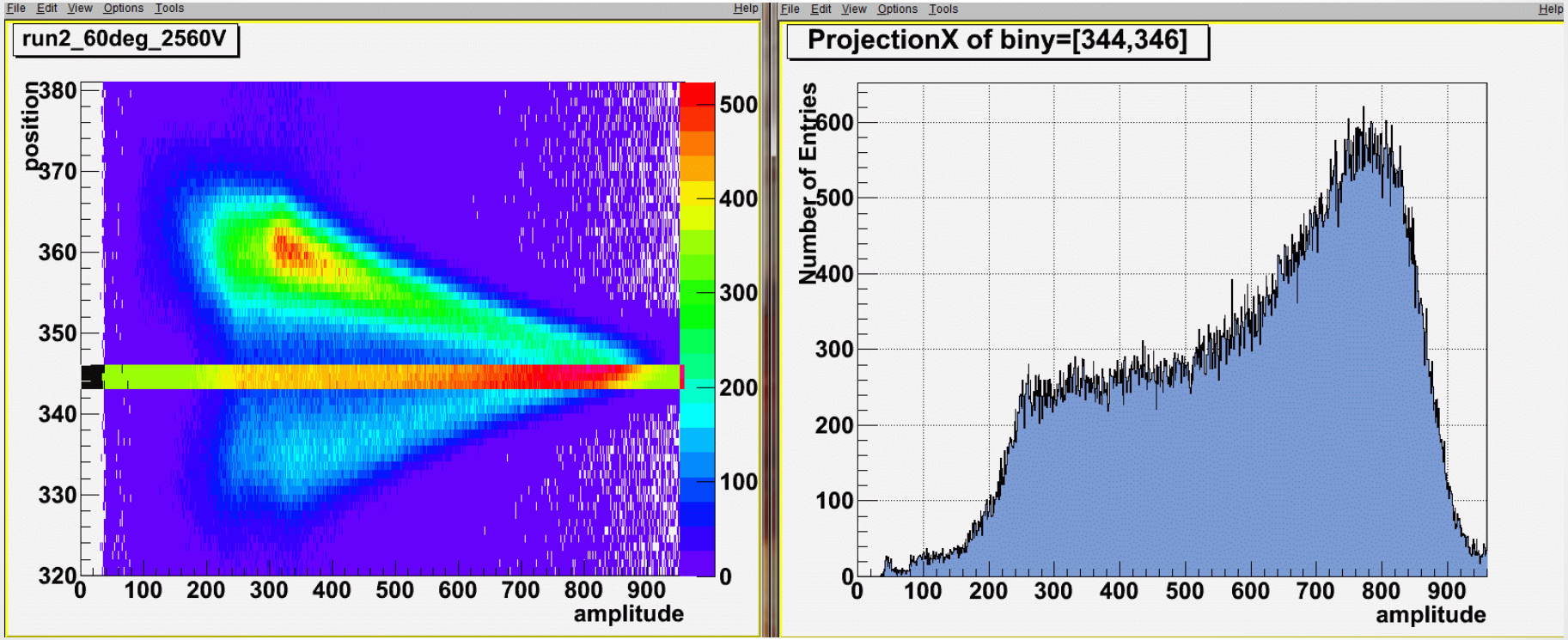


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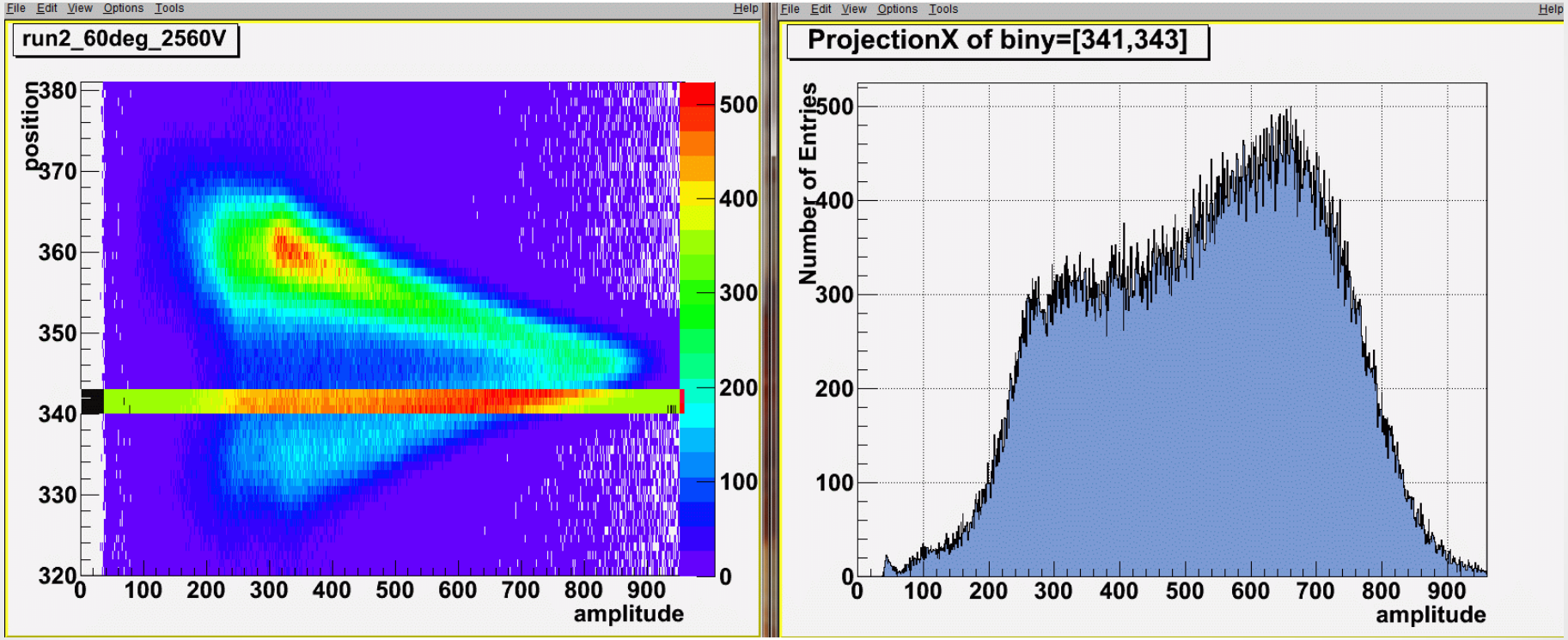


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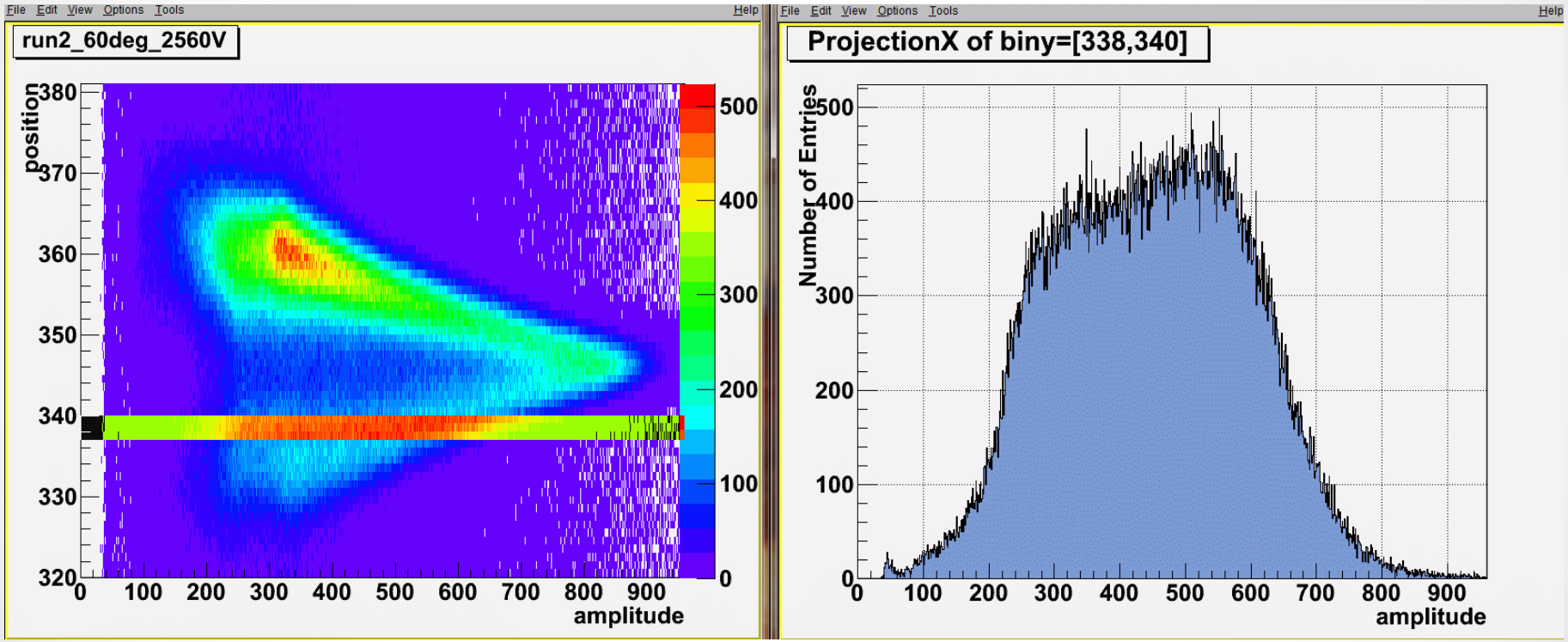


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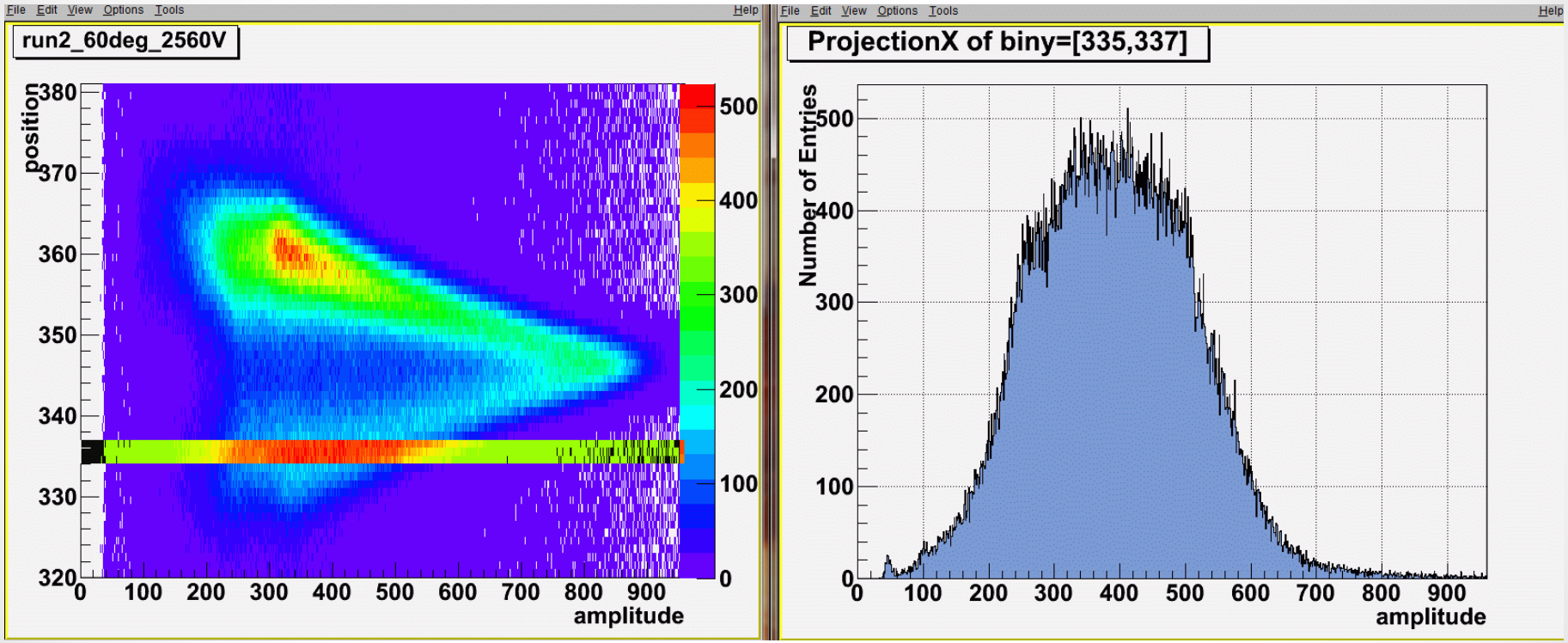


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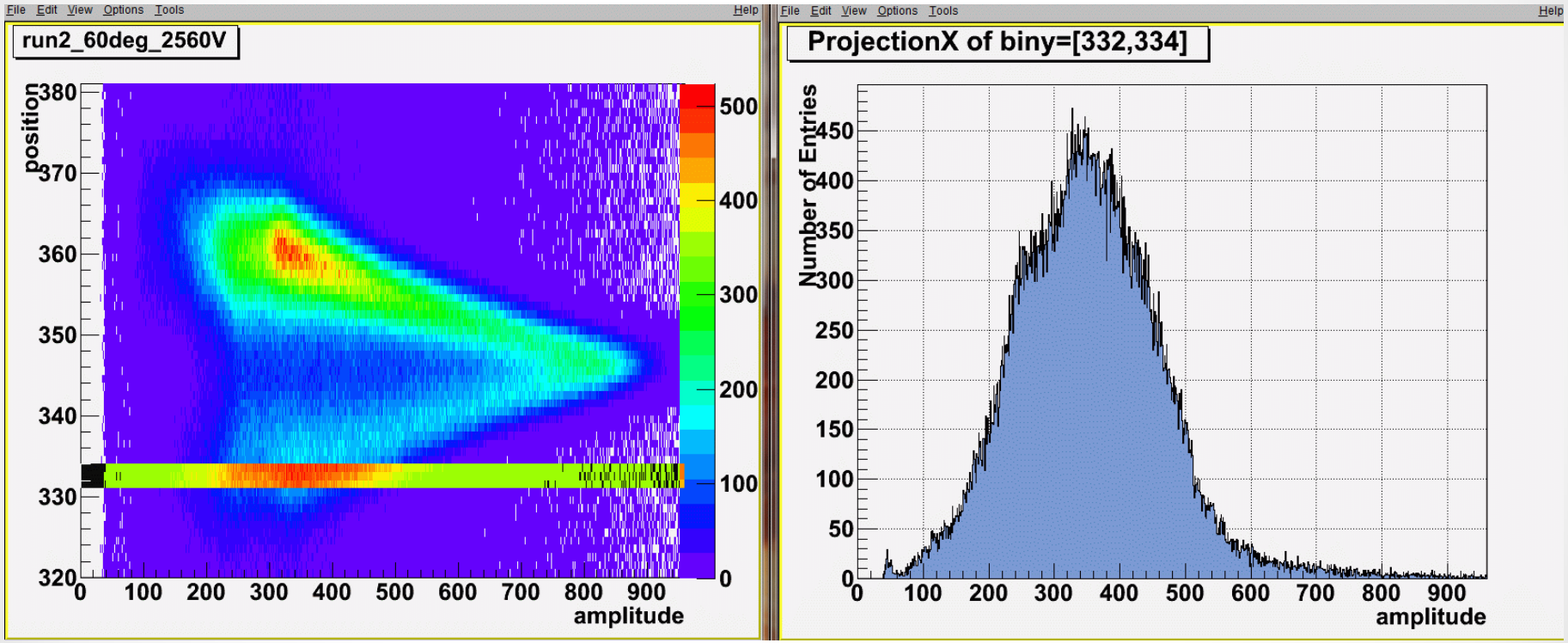


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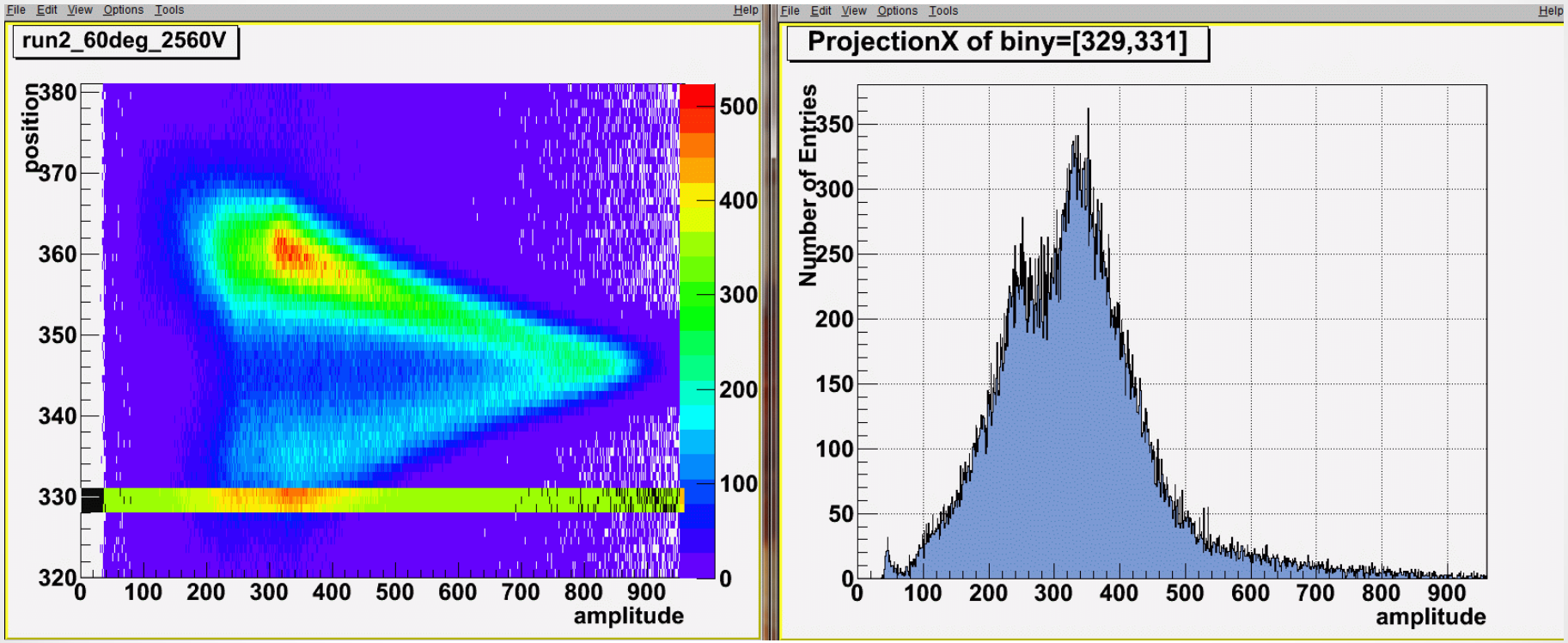
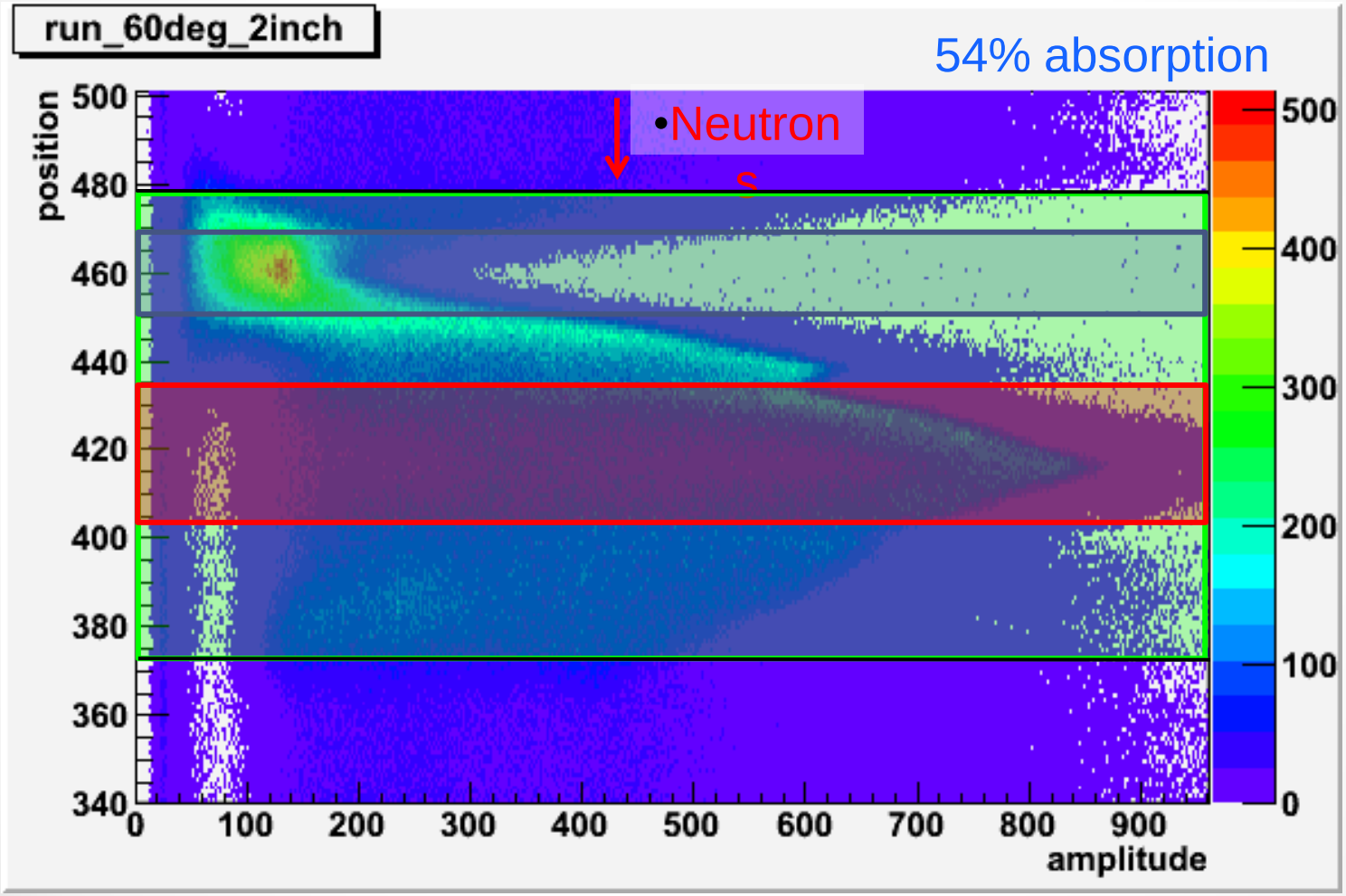
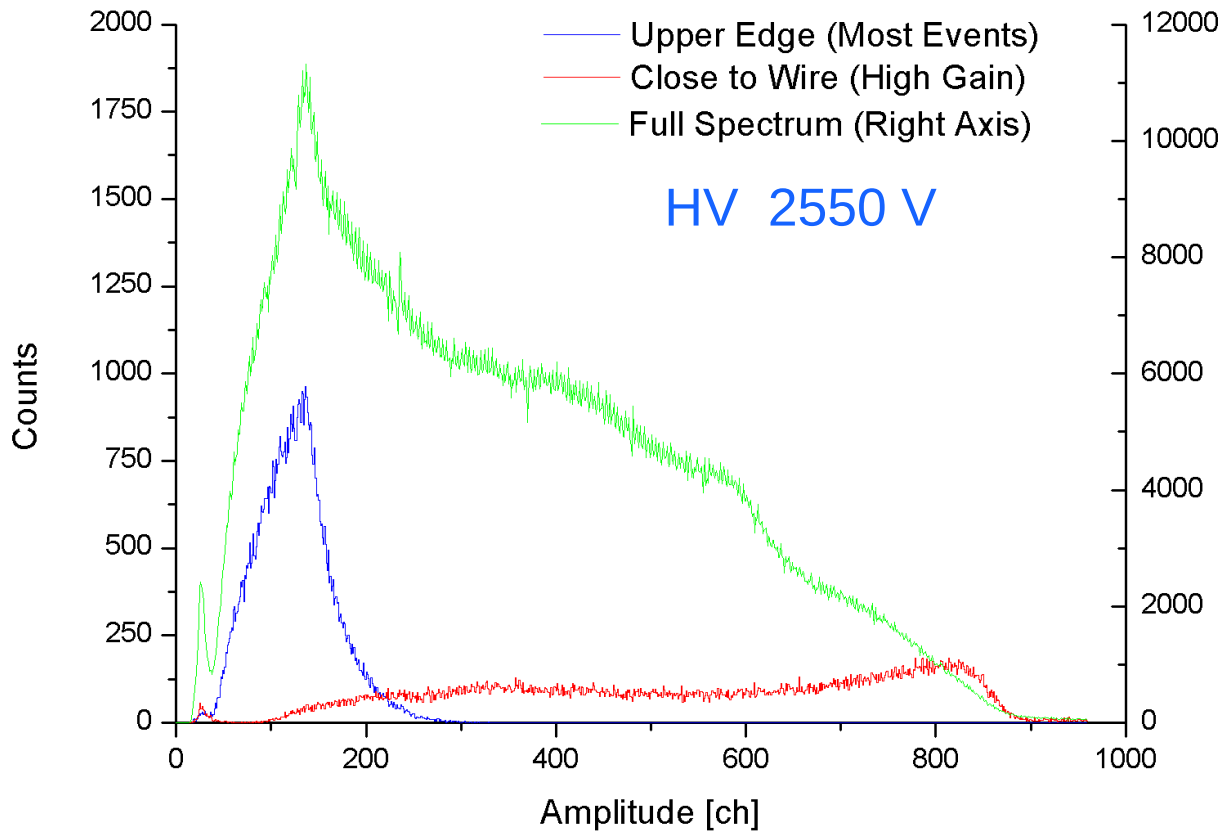



HV 2560 V, $\Delta x$ = 8 mm fwhm

# $BF_3$ – Pulse Height (2”)



# $BF_3$ – Pulse Height (2″)




**35 year old tube: $e^-$ attachment very likely**

# $BF_3$ – Sources of Attachment

*"…the main problem in the design of a good proportional counter is the purity of the gas"*

Thanks to N. Johnson (GE/RS)

THE REVIEW OF SCIENTIFIC INSTRUMENTS VOLUME 31, NUMBER 5 MAY, 1960

**Effect of Gaseous Impurities on $BF_3$ Proportional Counters***

J. Davila Aponte† and S. A. Korff
*Physics Department, New York University, New York 53, New York*

(Received December 17, 1959; and in final form, February 11, 1960)

1960

The effect which $SiF_4$, $SO_2$, and $SF_6$ have on the plateau and the pulse size distribution of a proportional counter were investigated. $SiF_4$ was tested at three pressures, 30, 45, and 60 cm, of $BF_3$ and its effect was found to be independent of the counter pressure for the range of value considered. From the variation in the plateau, the attachment probability for $SiF_4$ was calculated to be $h=1.485\times10^{-5}$ and its cross section for attachment to be $\sigma_a=5.12\times10^{-20}$ cm². The amount of these gases permitted, without the counter being affected beyond the limits of tolerance which are set up, were found to be 0.04% for $SiF_4$, 0.01% for $SO_2$, and $2.0\times10^{-6}$% for $SF_6$.

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$SiF_4$ 400 ppm

$SF_6$ 20 ppb

# $BF_3$ /$^{10}BF_3$ – Quality Today

2010

Two suppliers: (Ceradyne, Inc) Spectra Gases (Linde)

Natural $BF_3$

**Gasgemisch** mit folgender Zusammensetzung :

| Bestandteile | | Sollwerte | Einheit |
|---|---|---|---|
| BORTRIFLUORID E.G. | | Rest | |
| KOHLENDIOXID | $CO_2$ | < 25 | Vol-ppm |
| KOHLENMONOXID | CO | < 2 | Vol-ppm |
| SAUERSTOFF | $O_2$ | < 5 | Vol-ppm |
| SCHWEFELDIOXID | $SO_2$ | < 20 | Vol-ppm |
| STICKSTOFF | $N_2$ | < 15 | Vol-ppm |
| SULFATE | | < 8 | Vol-ppm |
| SILICIUMTETRAFLUORID | $SiF_4$ | < 20 | Vol-ppm |

Air Liquide

Electronegative gases play negative role

$^{10}$Boron Trifluoride

**Neutron Detector Grade**

Specification

| | | |
|---|---|---|
| 10Boron enrichment | 99.9 atom% | MS |
| Boron Trifluoride | 99.99% | |
| Nitrogen | < 15 ppm | GC |
| Oxygen + Argon | < 15 ppm | GC |
| Carbon Dioxide | < 15 ppm | GC |
| Carbon Monoxide | < 1 ppm | GC |
| Methane | < 1 ppm | GC |
| Hydrogen Fluoride | < 1 ppm | FTIR |
| Sulfur Dioxide | < 10 ppm | FTIR |
| Silicon Tetrafluoride | < 15 ppm | FTIR |
| Water | N.D.< 1 ppm | FTIR |

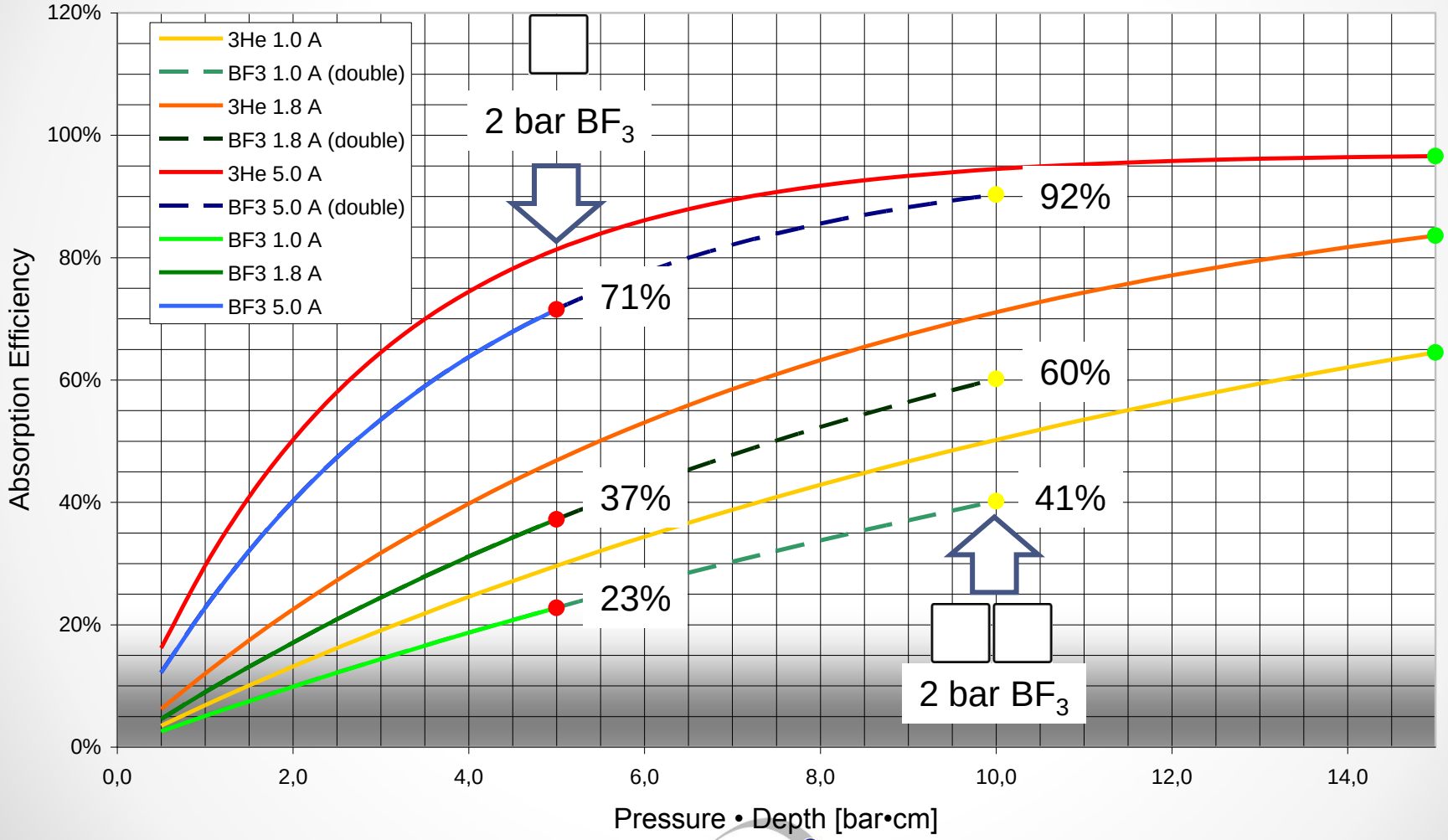
… no values below 1 ppm but interesting for $SF_6$

**BASF AG patented new technique for $BF_3$ production**

***DE3700244A1 21.07.1988***

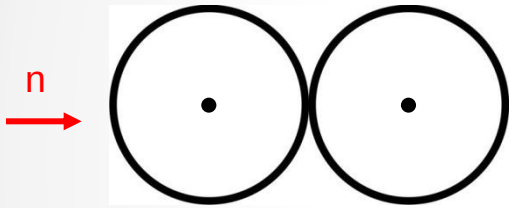
***EP0279438 08.05.1991***

# Efficiency – $^{10}BF_3$ vs. $^{3}He$



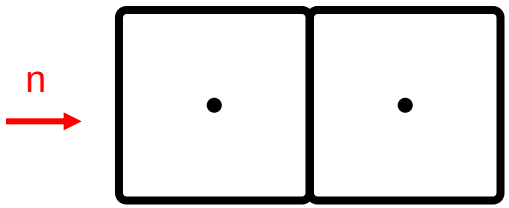
# $BF_3$ Tubes - Basic Geometries

2 x Ø 2.5 cm



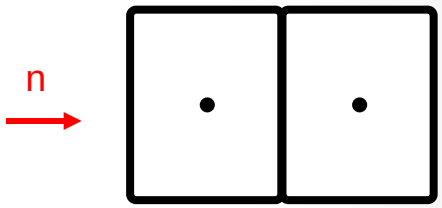

Pressure **2.6** bar
Efficiency@5.00 Å 90%

2 x 2.5 x 2.5 cm²




Pressure **2.0** bar
Efficiency@5.00 Å 90%

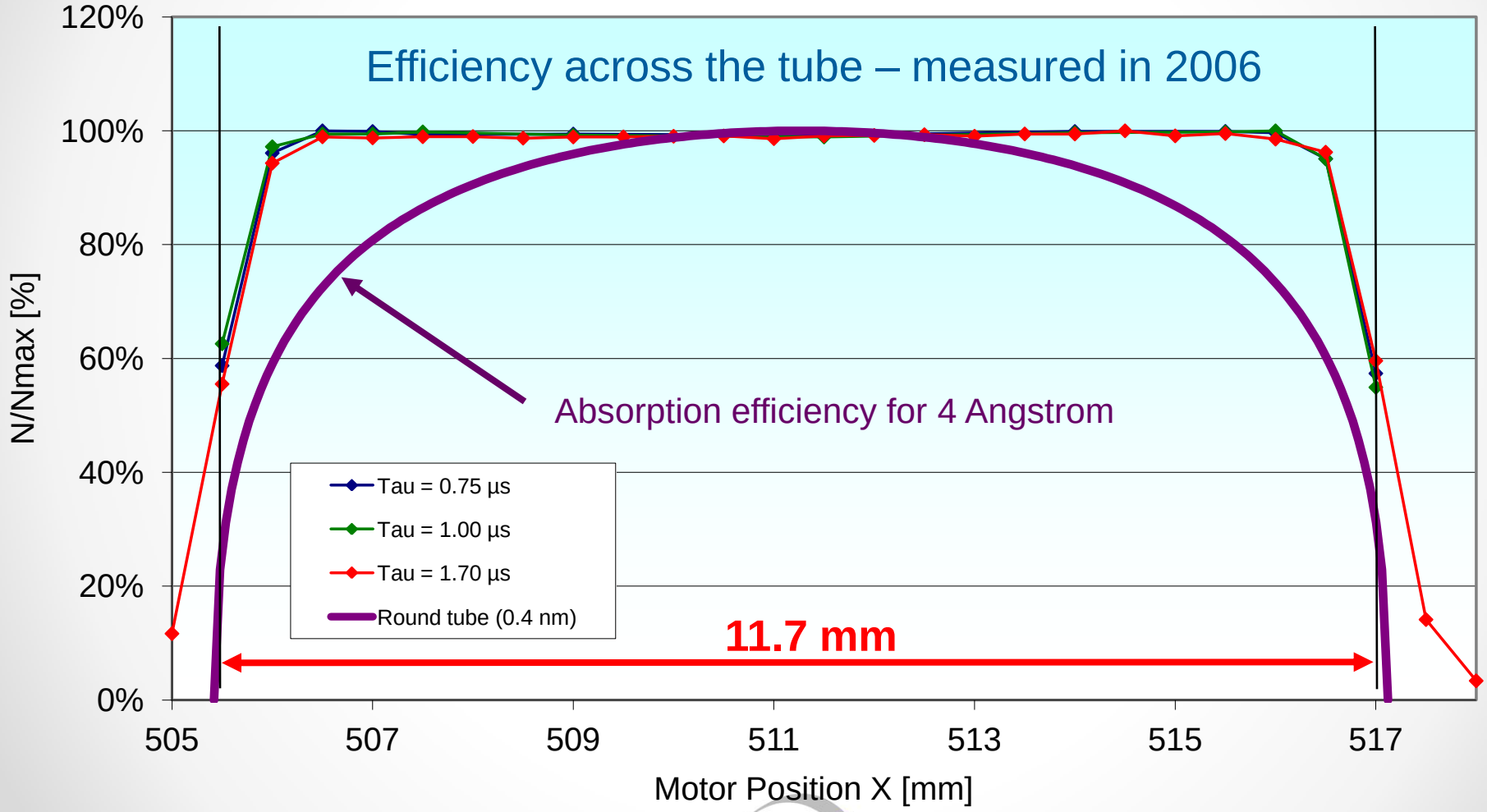
2 x 2.0 x 2.5 cm²



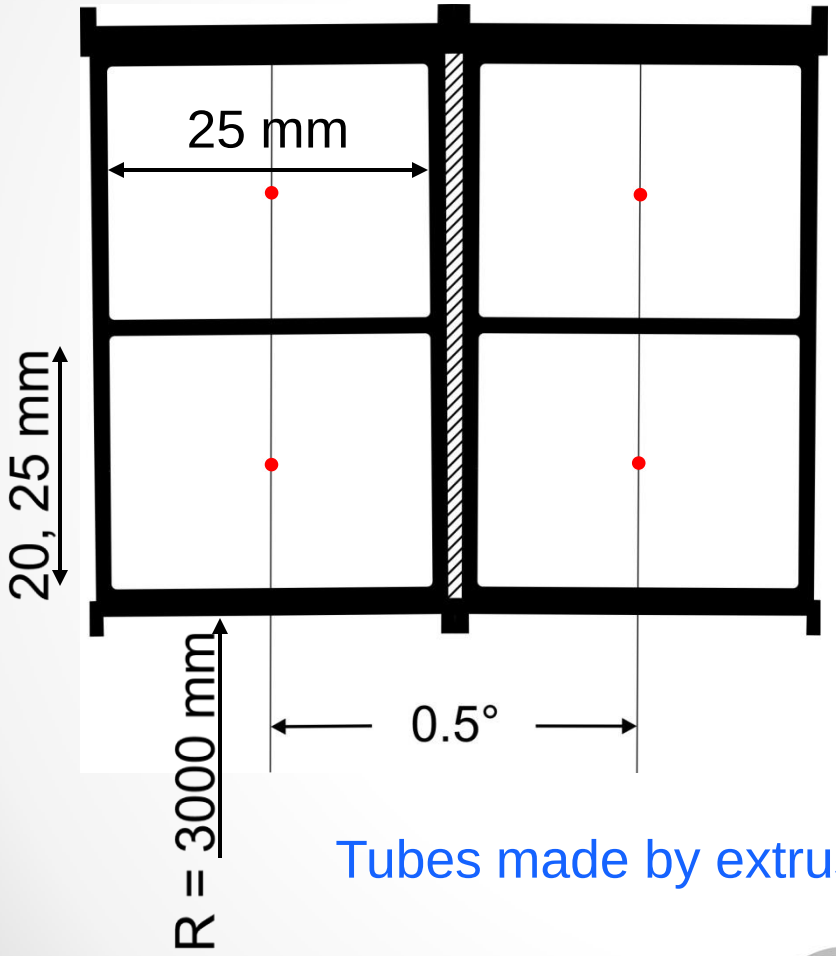

Pressure **2.5** bar
Efficiency@5.00 Å 90%
better ToF resolution

Can square tubes work at all?

# Performance of $^3$He Square Tubes (EXED)



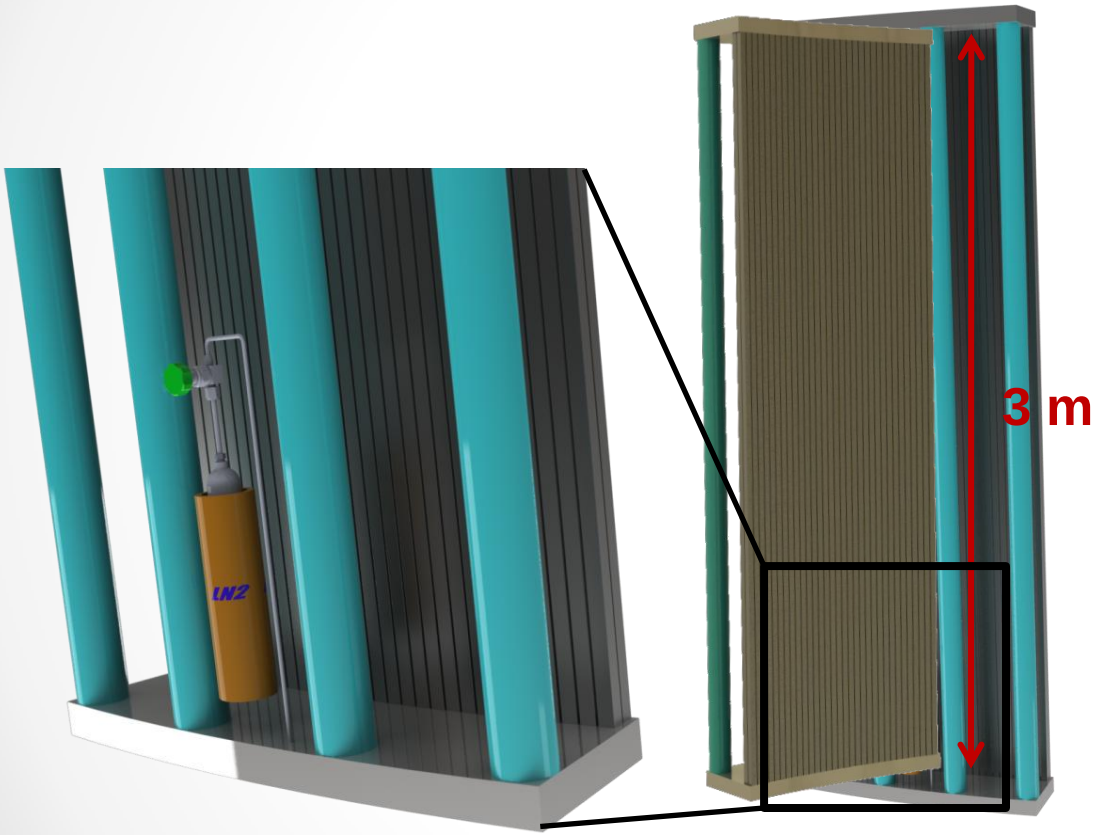
# Concept for NEAT

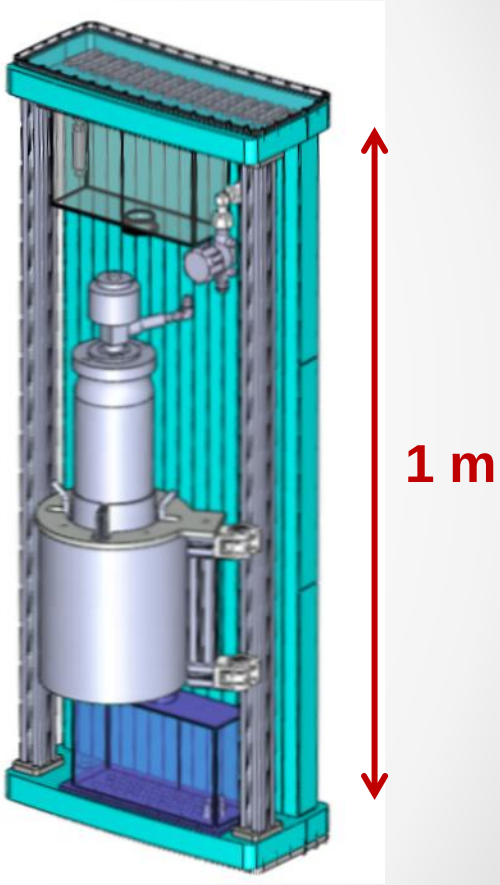



Tubes made by extrusion

- Extrusion: Well-known technique
- Straight profiles over meters
- Wall thickness 0.6 – 3.0 mm (depending on aspect ratio)
- Low costs for tools (2.5 k€) and material (3-10 €/kG)
- Many companies on the market

# Concept for NEAT

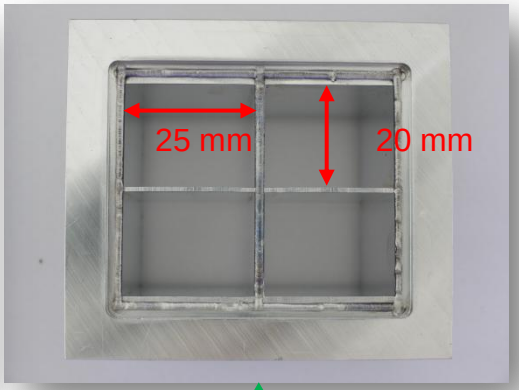


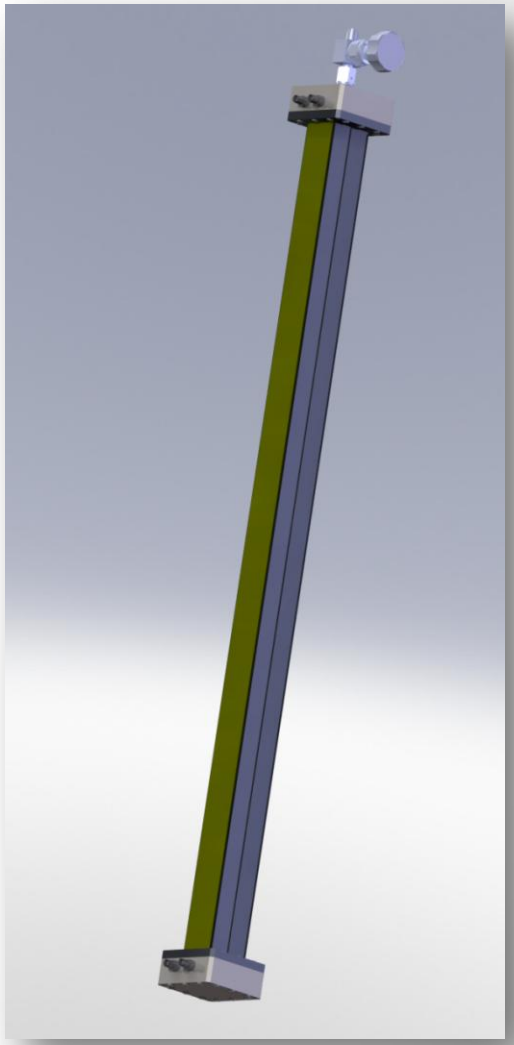

***based on concept of*** NEUTRONS FOR SCIENCE

# Concept for NEAT

Extruded Al 6060, 3.2 m long TwinTubes, 2x (20 x 25) mm²




neutrons



Tubes: company MIFA, www.mifa.nl

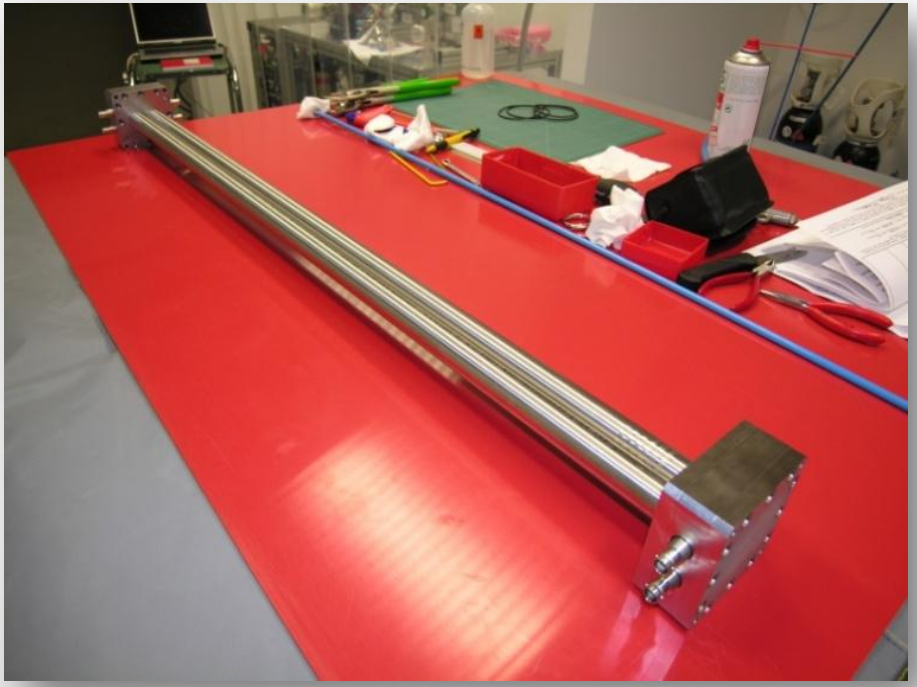
3 kinds of coating (no, Ni, Surtec)

Laser welding: HZB Workshop, company DSI

Welding problem! (still to be solved)

# 1st $BF_3$-Prototype of HZB

Round tubes (SS)



What can be tested:

Variation of the anode wire diameter
Variation of $^{10}BF_3$ gas pressure/mixtures
Freeze out procedure

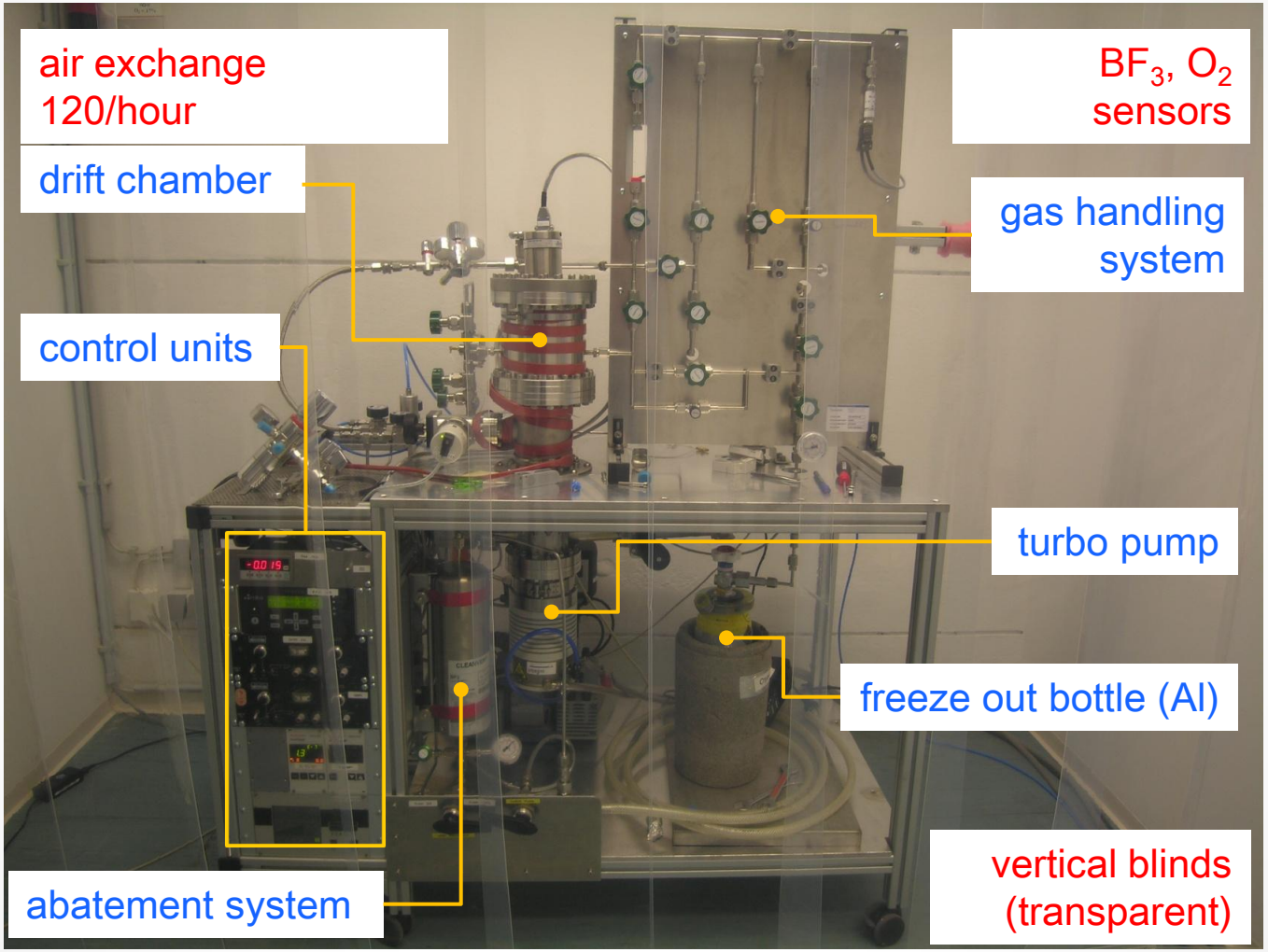
Welding: HZB Workshop, conventional method (material: SS 1.4435 (316L), 1” OD, 1.65 mm wall)

Filled in HZB‘s special laboratory
First tests with AmBe source

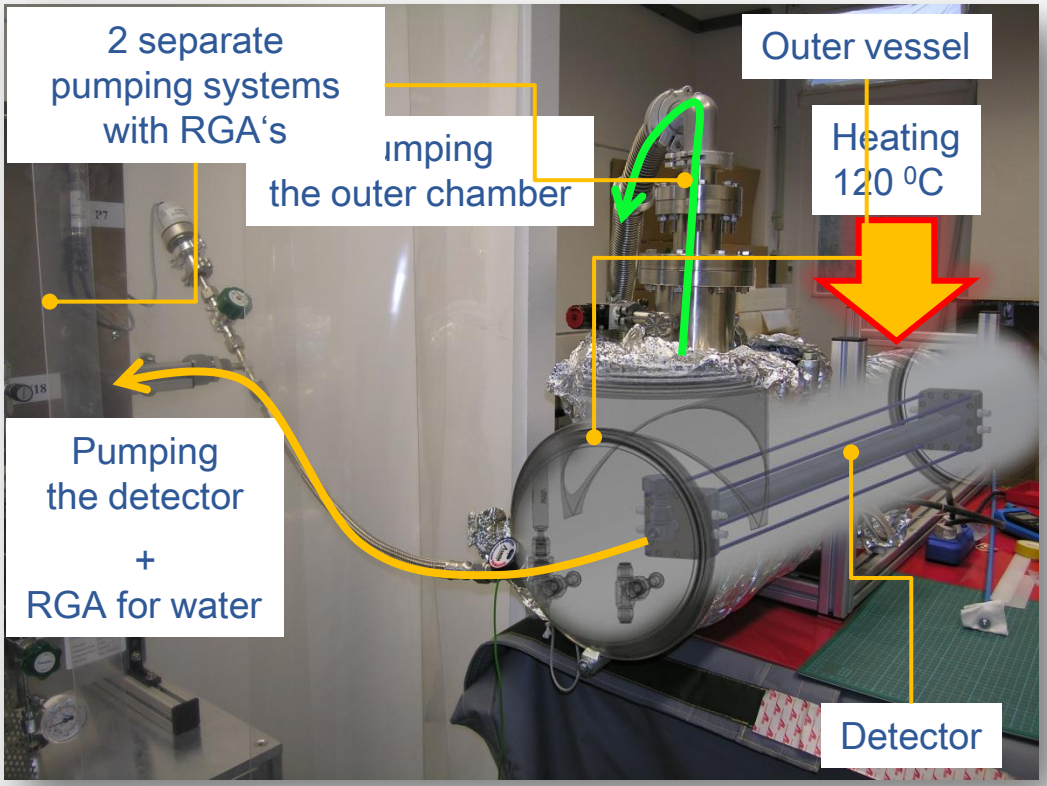
# $BF_3$-Laboratory Requirement

- Very good ventilation: 120 exchanges per hour
  - cost for ventilation system: reduce volume
  - monitoring of function: alarm for failure
  - exhaust connect to abatement system (hold full capacity)
- Continues monitoring of $BF_3$ level at several places
  - certified sensors
  - level ≥ 1ppm: alarm & escape
- Good working practices
  - use metallic seals
  - double-check tube connections, valves etc.
  - perform pre-tests with harmless test gas (similar properties)

# $BF_3$-Laboratory - Work in Safe Environment



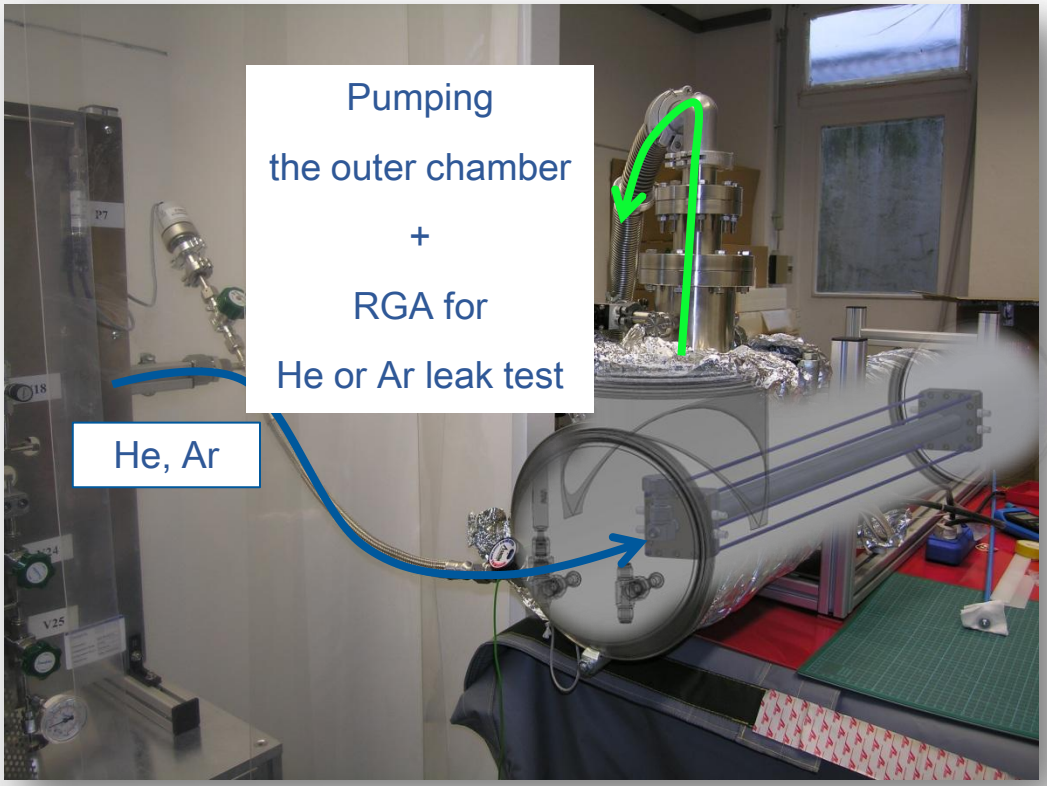
# $BF_3$-Filling Procedure 1




Rough vacuum test

Bake-out

# $BF_3$-Filling Procedure 2

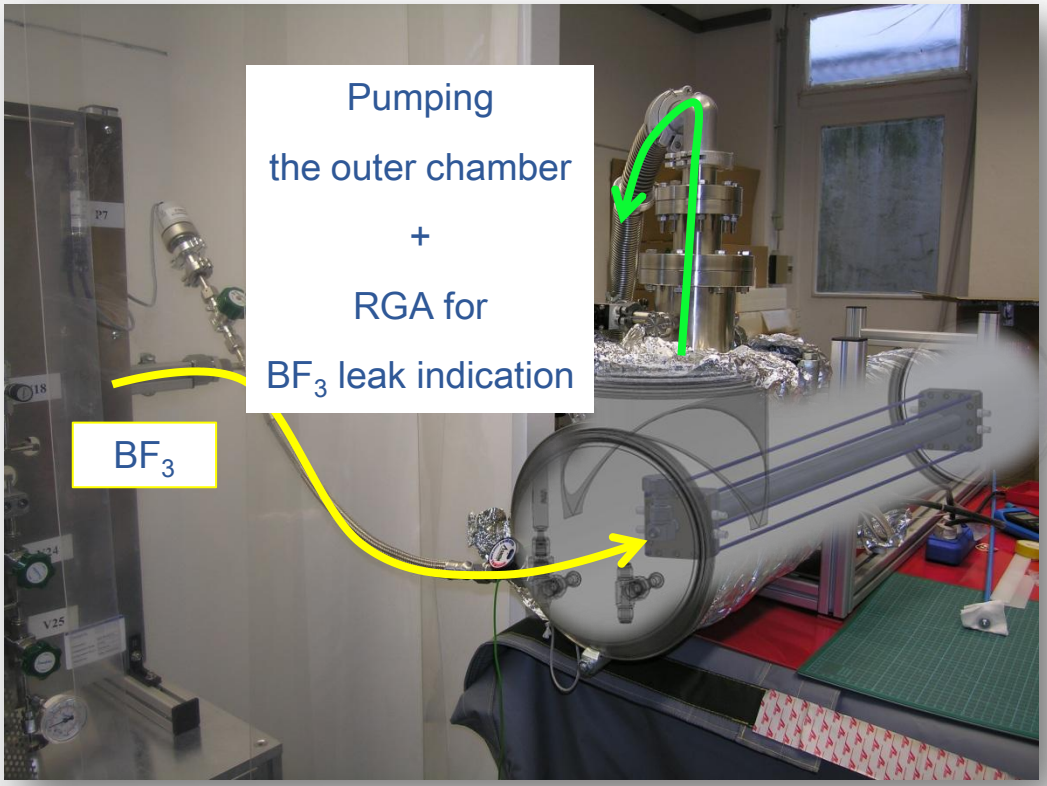



Rough vacuum test

Bake-out

He overpressure test

# $BF_3$-Filling Procedure 3




10-14 days for one detector

Rough vacuum test

Bake-out

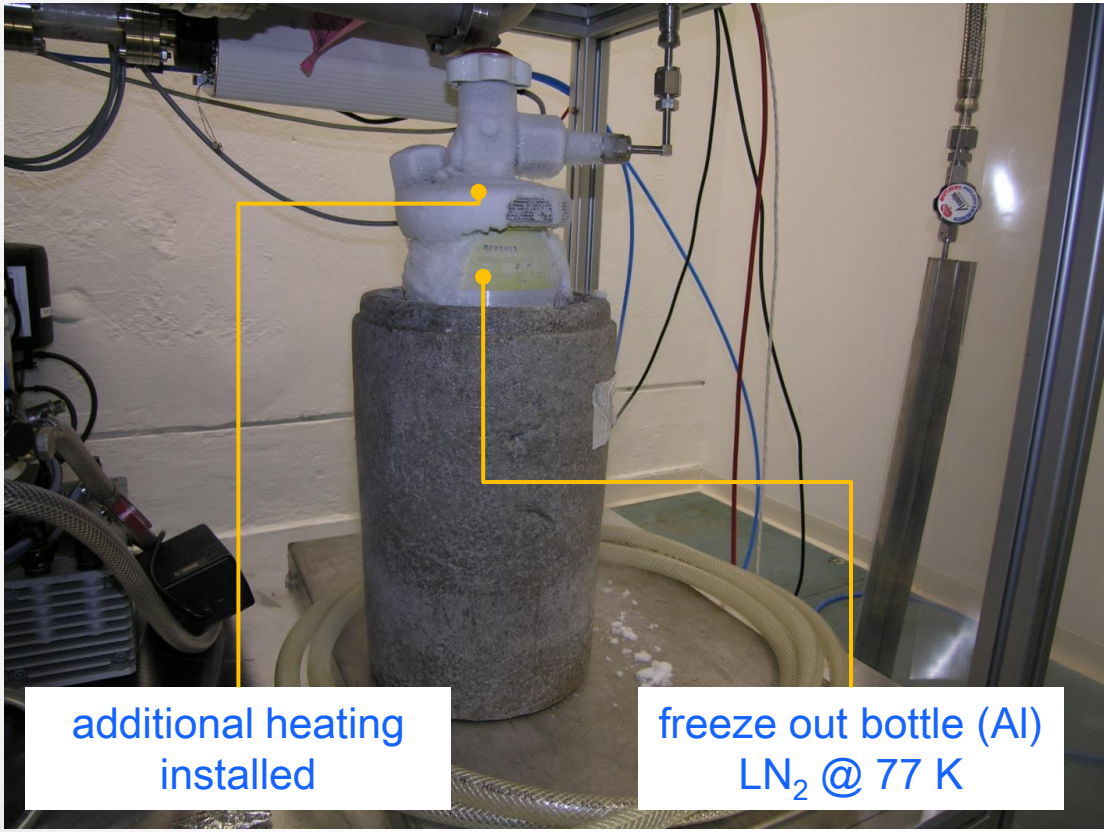
He overpressure test

Filling the detector with $BF_3$

In any case the gas stays inside the vessel!

New safety feature

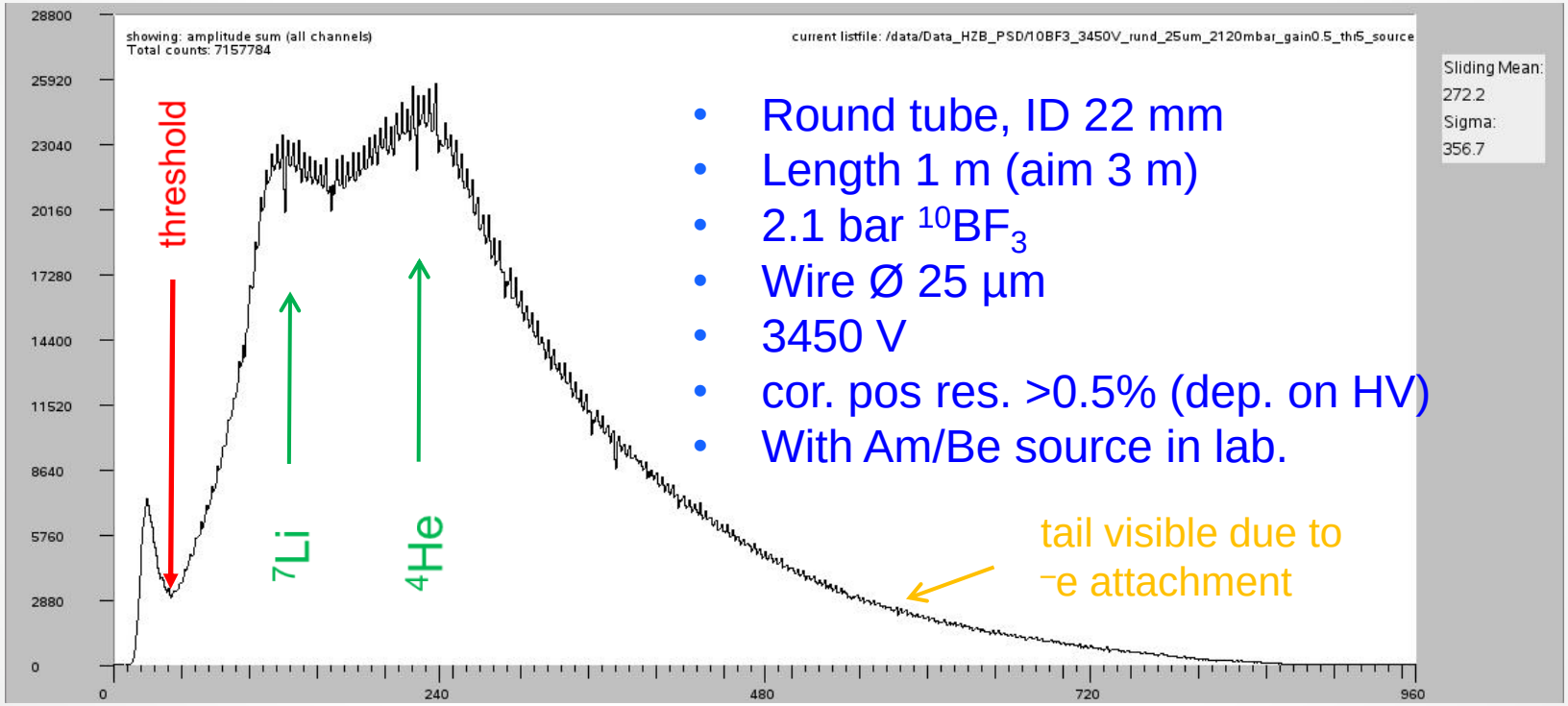
# Safe Dumping of $BF_3$




- Pre-tested with $CF_4$
- $BP_{CF4}$ = 145 K
- $BP_{BF3}$ = 173 K
- Few minutes < 1mbar
- Rest: flushing with $N_2$ & pumped through abatement system

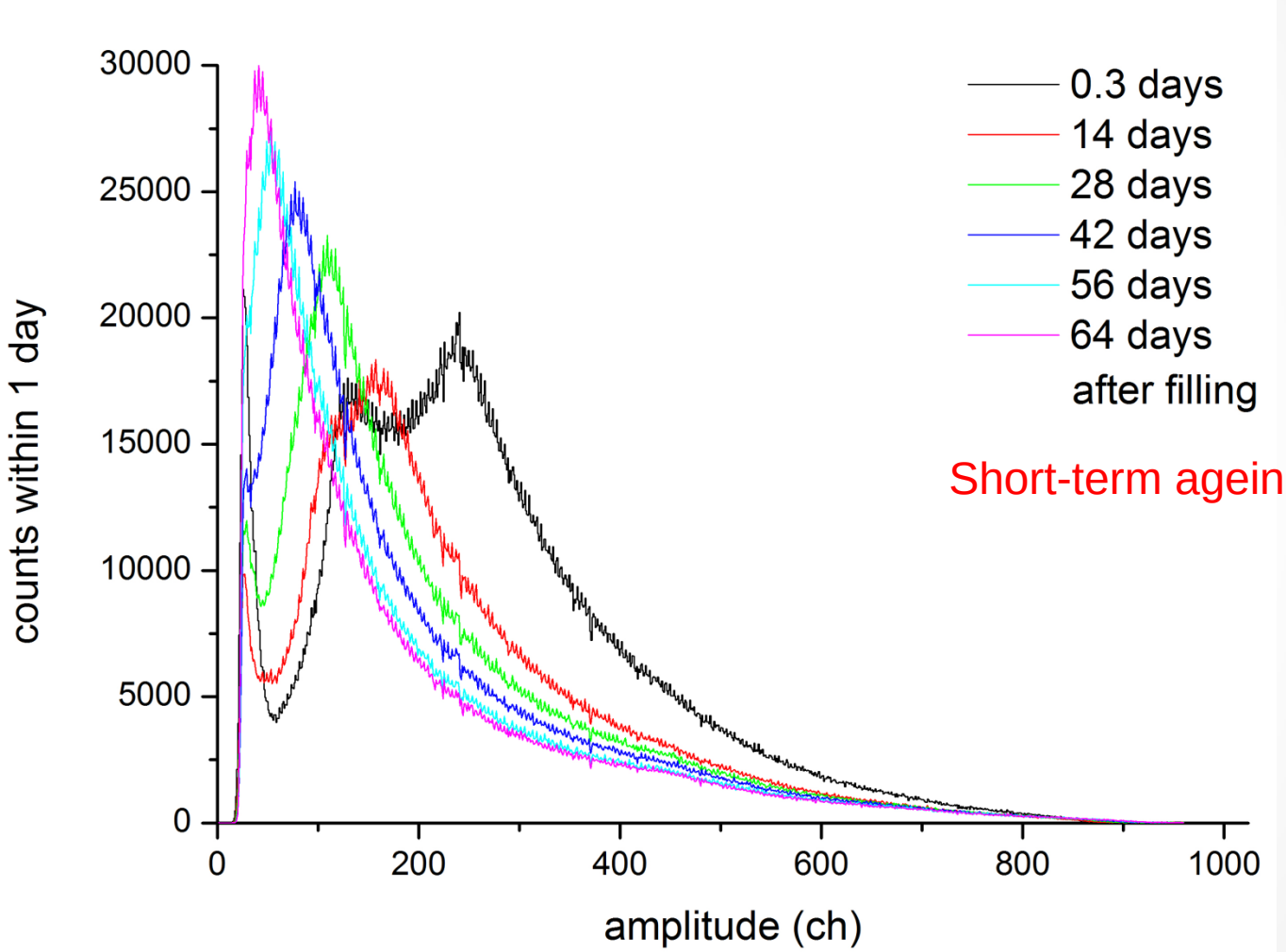
# HZB $^{10}BF_3$ Tube – Pulse Height

$^{10}B(n,^4He)^7Li$




- Round tube, ID 22 mm
- Length 1 m (aim 3 m)
- 2.1 bar $^{10}BF_3$
- Wire Ø 25 µm
- 3450 V
- cor. pos res. >0.5% (dep. on HV)
- With Am/Be source in lab.

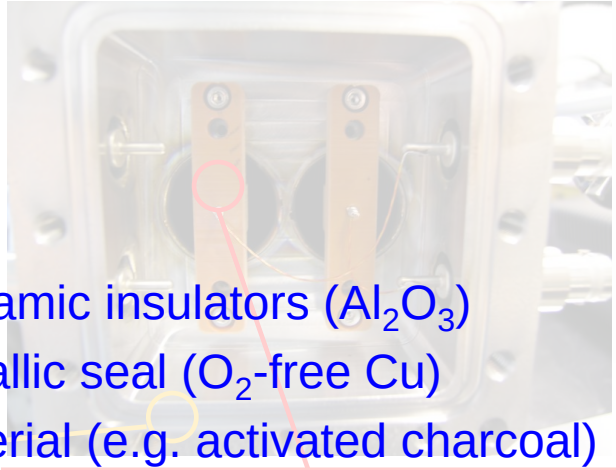
# HZB $^{10}BF_3$ Tube – Signs of Ageing




Short-term ageing!

# Any Idea?

fresh PEEK after installation



PEEK after $BF_3$

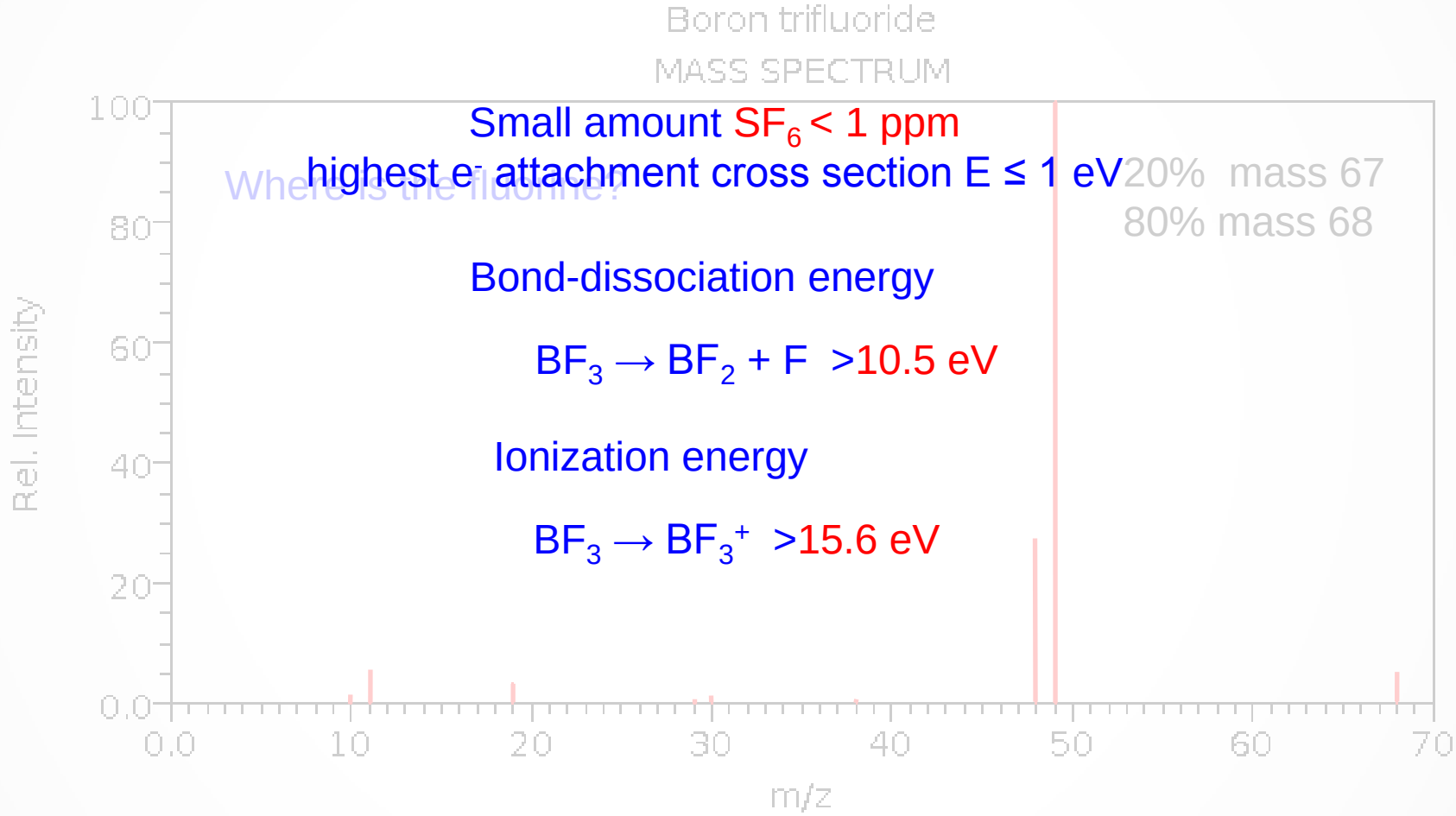
- Replacing PEEK by ceramic insulators ($Al_2O_3$)
- Replacing Viton by metallic seal ($O_2$-free Cu)
- Fluorine absorbing material (e.g. activated charcoal)

dry droplets inside Viton O-ring

few hours after opening: droplets on surface

0.5 mm

# Reason For Ageing?




Small amount $SF_6$ < 1 ppm

highest $e^-$ attachment cross section E ≤ 1 eV

Bond-dissociation energy

$BF_3 \rightarrow BF_2 + F$ >10.5 eV

Ionization energy

$BF_3 \rightarrow BF_3^+$ >15.6 eV

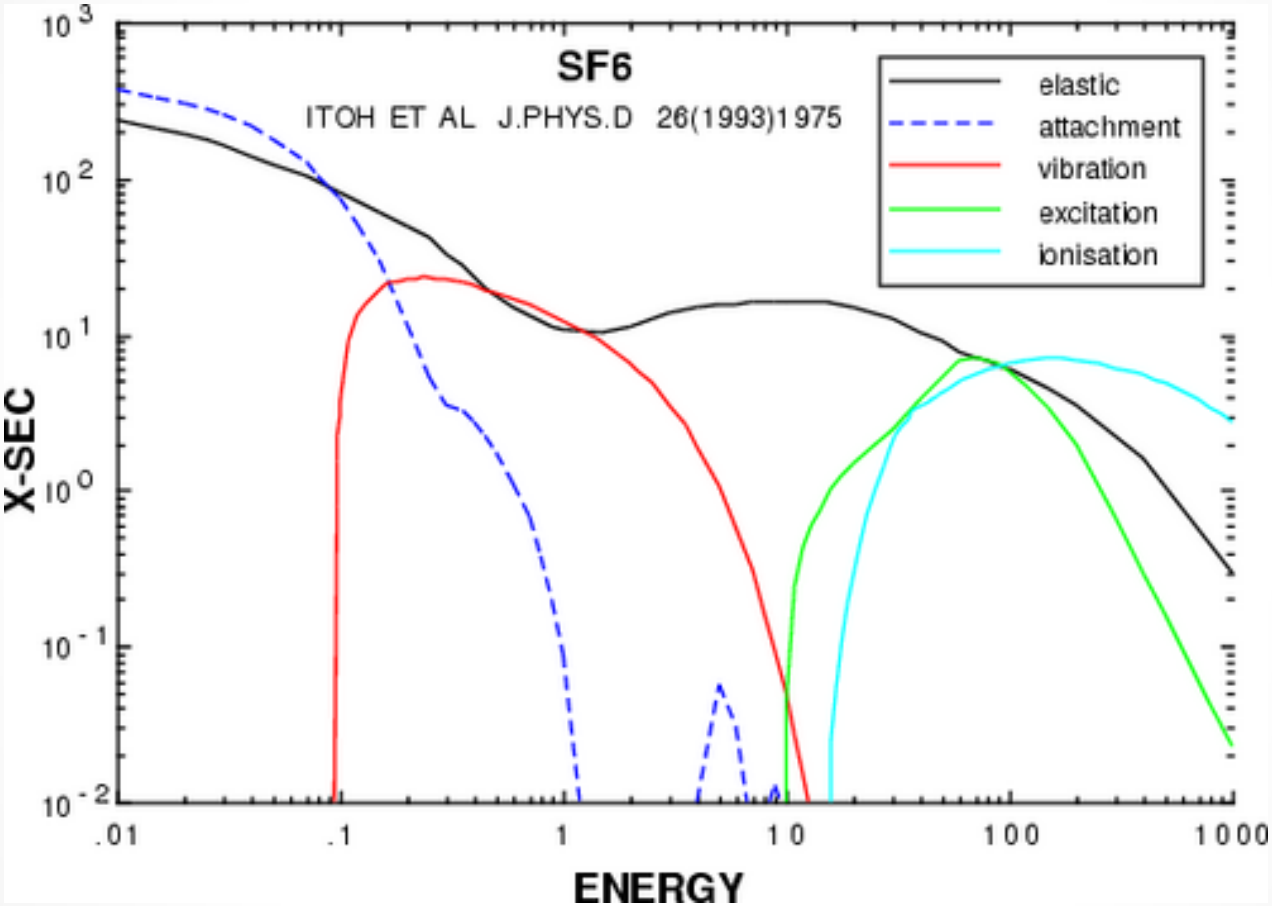
# Summary & Outlook

- Demonstrated LPSDs with $^{10}BF_3$ work at ~2 bar
  - same performance as $^{3}He$ – except efficiency → twin tube design
  - same electronics can be used
  - investigate 2.5 bar (better ToF res.)
- Ageing is an issue! → careful material selection (no PEEK!)
- Safety is important, in practice some effort – no hindrance
- Transportation of gas or detector is possible – some effort
- Large Special Laboratory at HZB ready in autumn 2012
- Larger prototype for NEAT (16 twin tubes, 1m long) to be built soon (welding!)
- Backup solution for in cooperation with ILL in preparation
  - (IN5-like, 3 consecutive round SS tubes)

# Thank you very much for your attention!

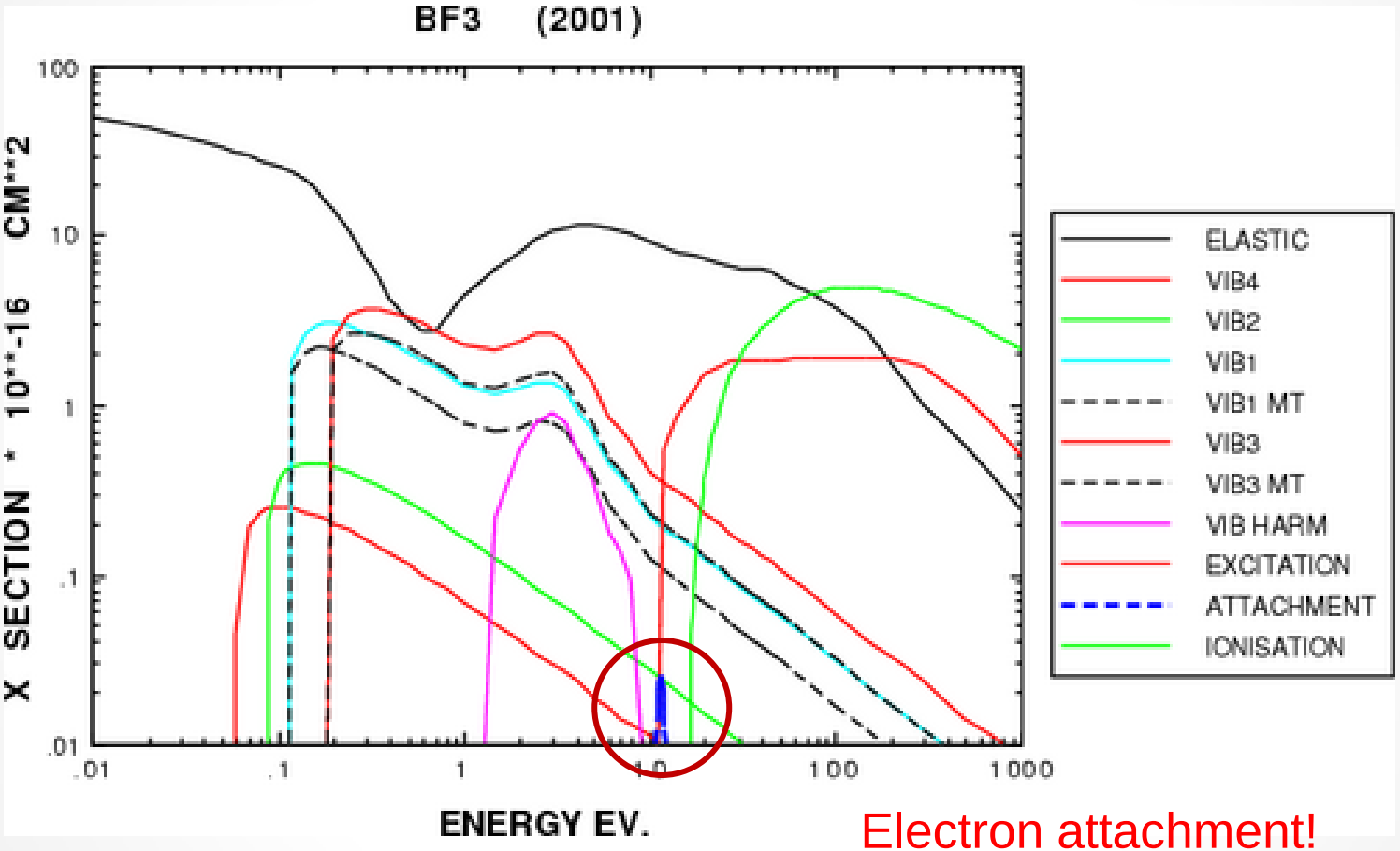
*The right choice of a transportation company is important!*
*Example of an expensive one.*

# Cross Sections - $SF_6$




source: http://garfield.web.cern.ch/garfield/help/

# Cross Sections - $BF_3$




Electron attachment!

source: http://garfield.web.cern.ch/garfield/help/