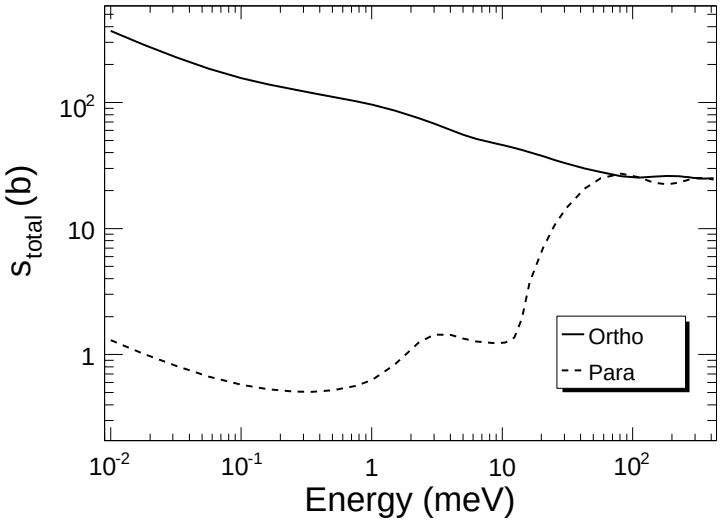
Measurement of the Liquid Hydrogen Ortho/Para Conversion Rate at the Manuel Lujan Jr. Neutron Scattering Center

M. Mocko, G. Muhrer

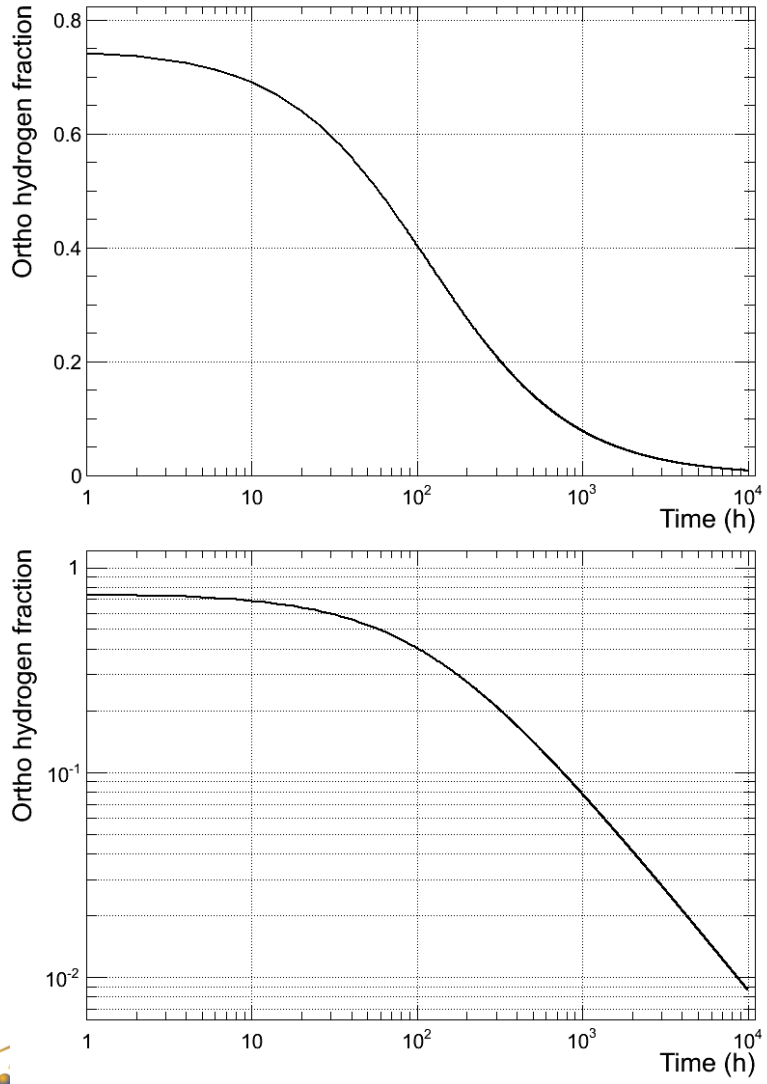
ICANS XX, Bariloche Argentina

Ortho/Para hydrogen

- **Two spin states of H_2 molecule**
  - Parallel (ortho) = spin 1
  - Anti-parallel (para) = spin 0
- **Different energy levels (14.7 meV)**
- **Radically different neutron scattering cross sections below 30 meV**
- **O/P=75/25 right after liquefied (or in gas form)**
- **O/P=0.2/99.8 in equilibrium (undisturbed)**
- **What about an operating LH_2 moderator?**



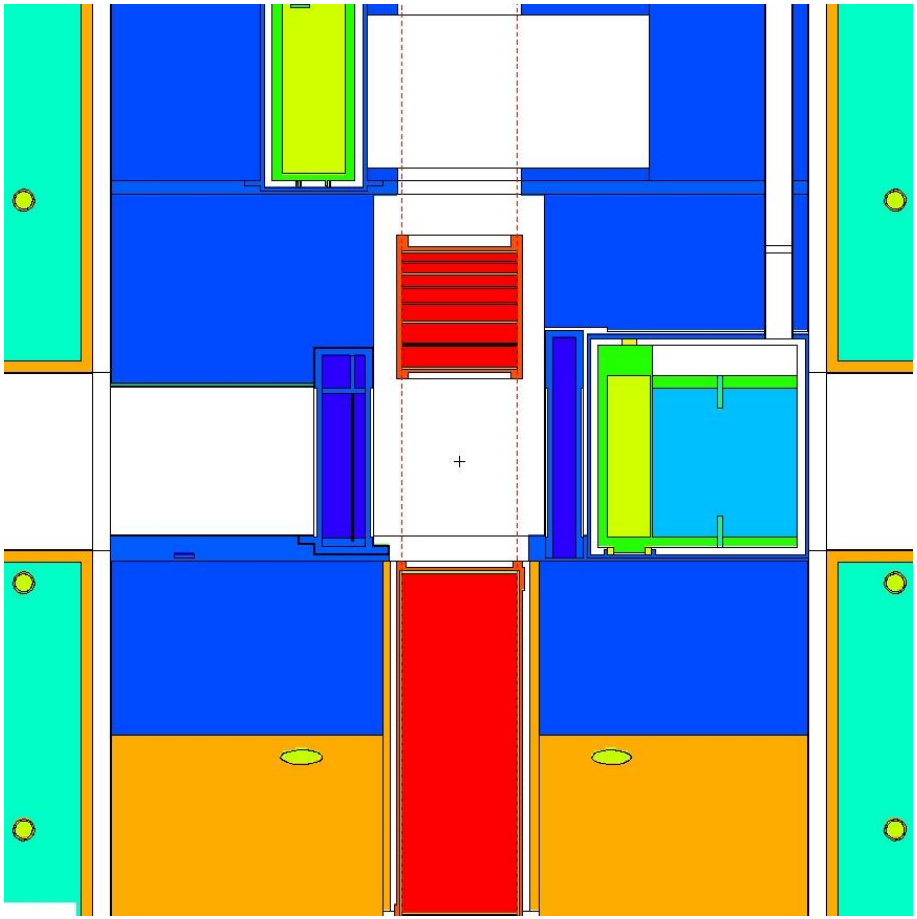
Natural Ortho-Para conversion



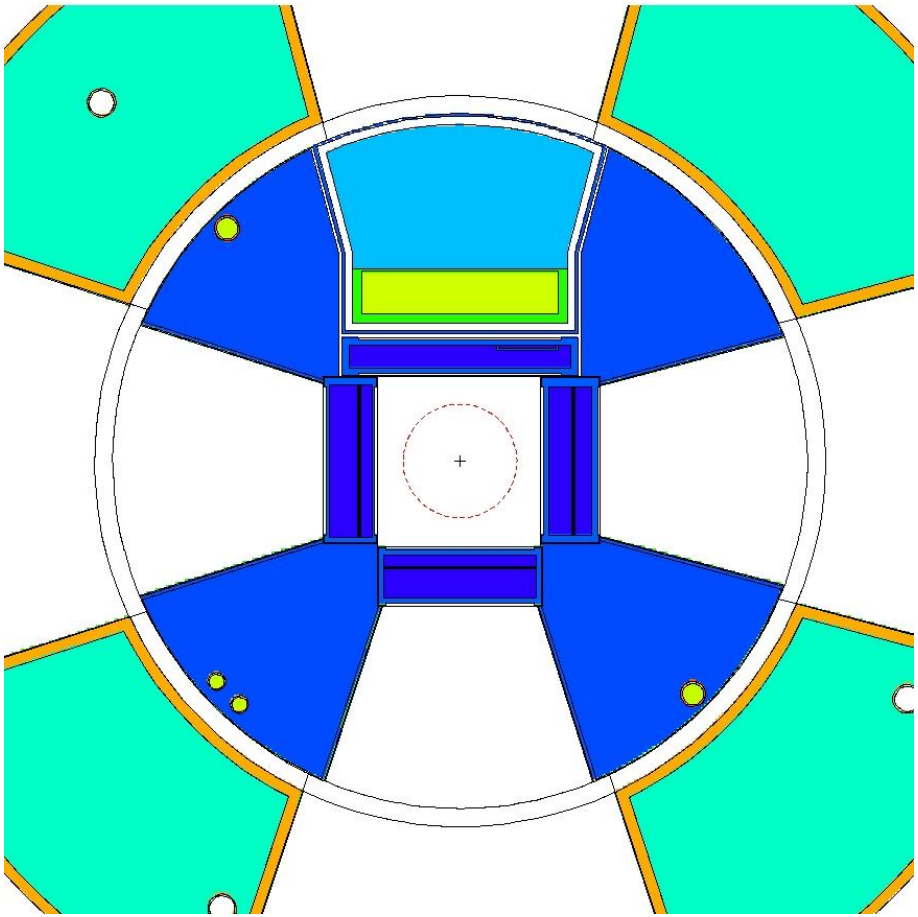
- **Natural conversion:**
  - O/P=75/25 (0 h)
  - O/P=50/50 (58 h)
  - O/P=25/75 (1.4 weeks)
  - O/P=10/90 (1 month)
  - O/P=1/99 (1 year)
- **Avoid changing neutron spectrum ➔ control the O/P fraction**
  - Use para-hydrogen (catalyst to speed up the conversion rate)
  - Lujan TMRS: O/P≥20/80!

Lower-tier LH_2 moderator with cold Be reflector/filter (Mark-III)

Elevation view:

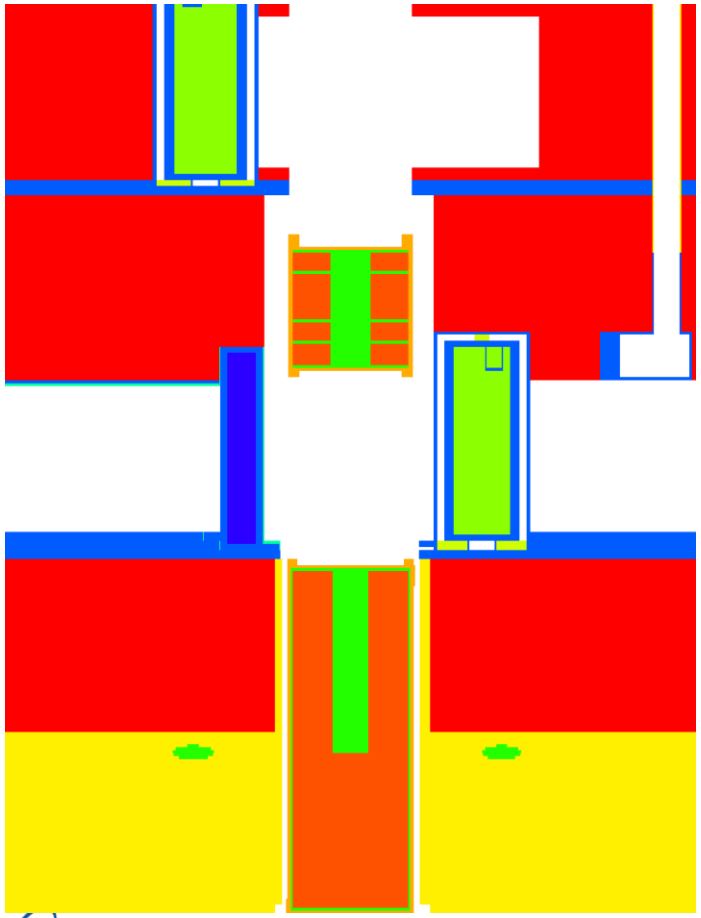


Lower-tier section:

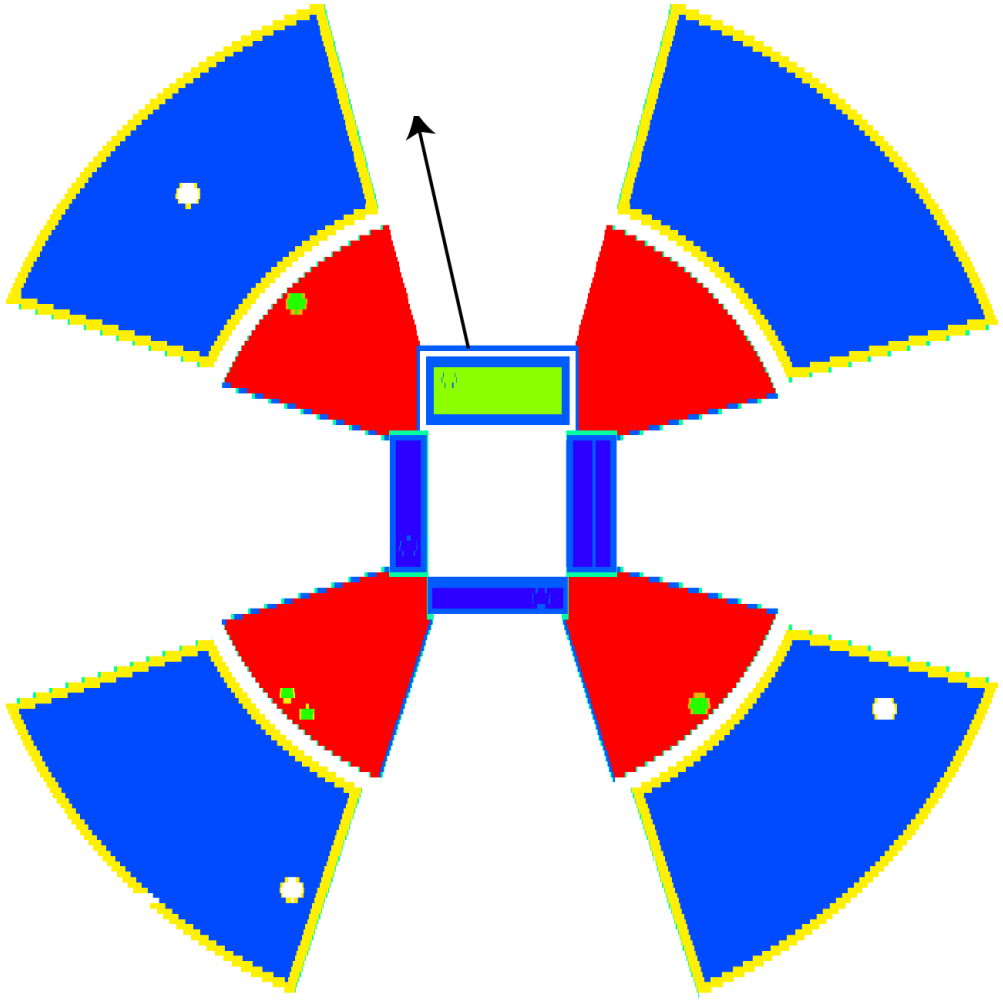


Lower-tier LH_2 moderator (Mark-II)

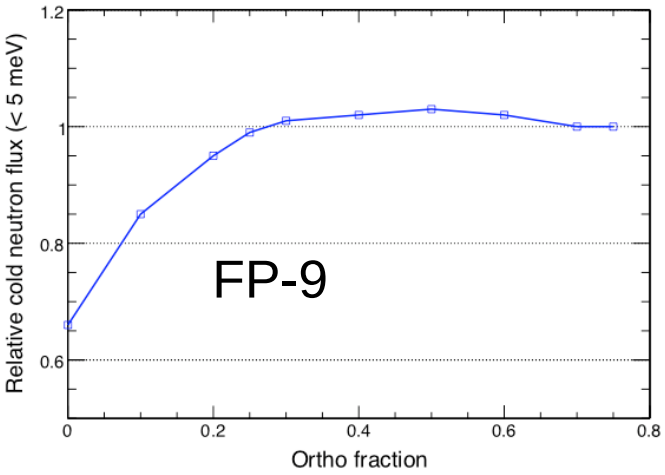
Elevation view:



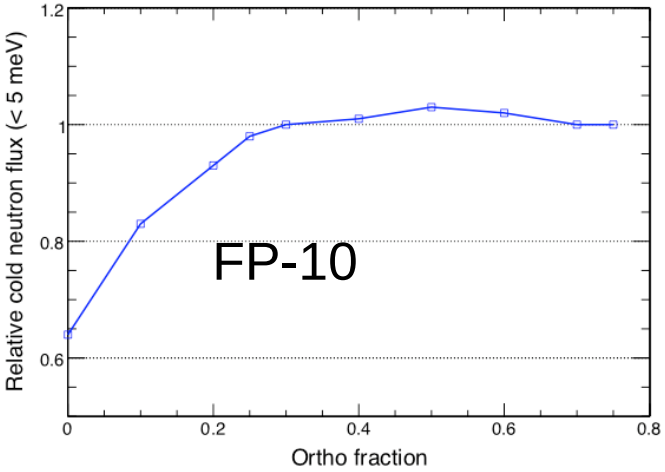
Lower-tier section:

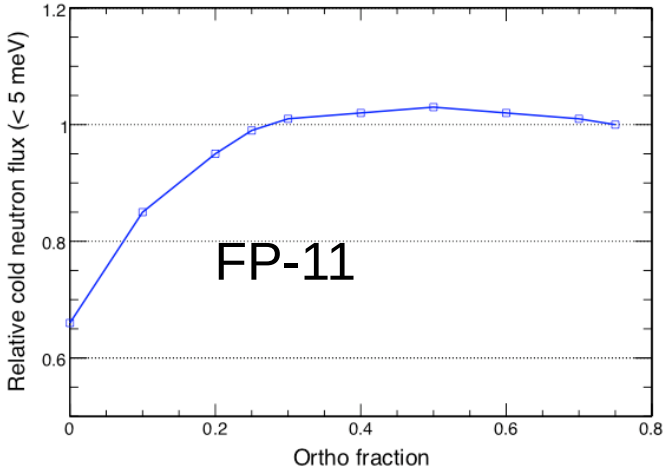


Ortho/para ratio dependence



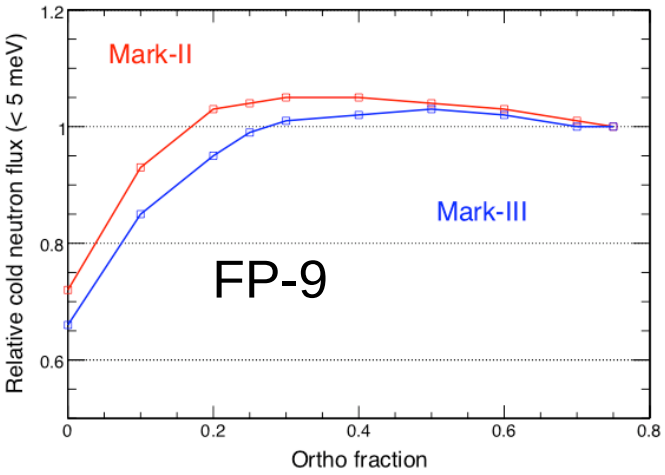
- From optimization study of the cold Be reflector-filter in Mark-III design
- Cold neutron flux changes as a function of ortho/para ratio (not surprising)
- Variations minimal if ortho fraction > 0.25



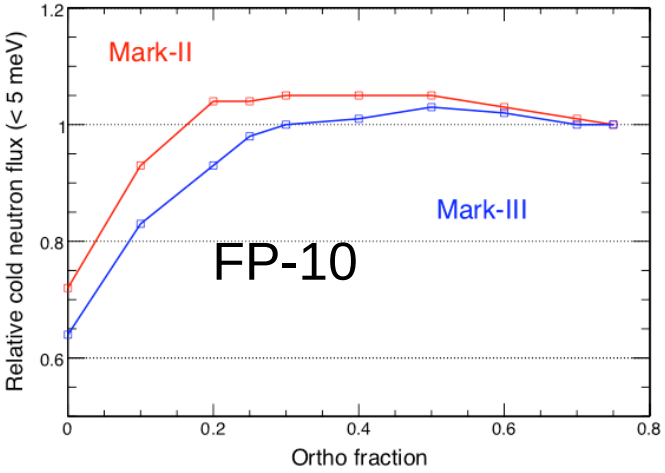


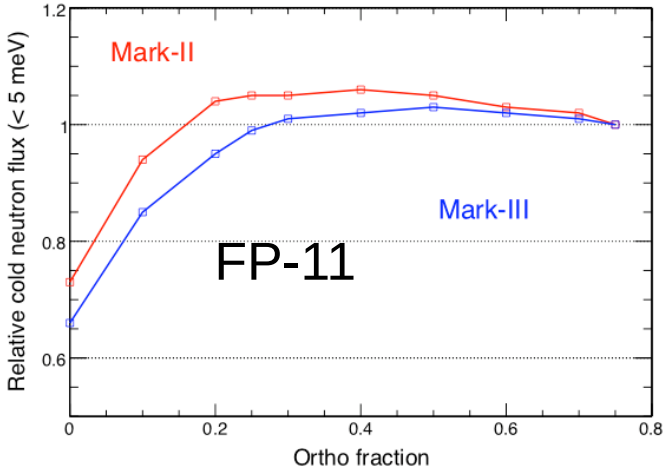
Mark-III:
partially coupled liquid hydrogen moderator with cold beryllium reflector-filter

Ortho/para ratio dependence



- From optimization study of the cold Be reflector-filter in Mark-III design
- Cold neutron flux changes as a function of ortho/para ratio (not surprising)
- Variations minimal if ortho fraction > 0.25
- Dependence qualitatively similar to Mark-II design





Mark-II:
partially coupled liquid hydrogen moderator

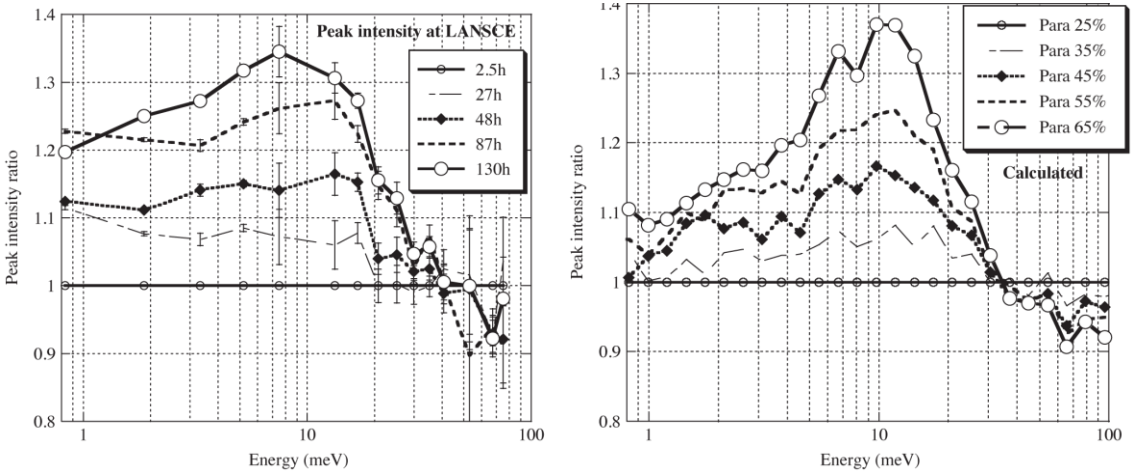
Measuring the O/P ratio

- **Direct measurement**
  - Measuring the ortho/para content in the liquid hydrogen while in operation
  - Extremely challenging
  - Preferably in moderator volume (harsh radiation environment)
  - First attempts at Lujan Center:
    - Installation of a Raman spectrometer with long fiber optical leads
    - Need to sample the hydrogen under scrutiny (measurement in vent line)
    - *No measurements done yet*

- **Indirect measurement**
  - Measuring the consequences of the ortho/para conversion in the liquid hydrogen
  - Long term measurement of the neutron beam characteristics (flux, energy spectrum, time emission spectra)
  - Challenges:
    - Ensuring stability of the experimental equipment over long periods of time
    - Availability of a beamline for such measurements
    - *Tying the neutron signature to O/P observable*

Experiments with Mark-II

- Measurement of neutron energy and time emission spectra
- Experiment 132 hours (O/P=35/65)



M. Ooi et al., NIMA 566 (2006) 699-705

Conclusions:

"From the comparison of the change of spectral intensity and the pulse shape with the calculated ones, we estimated the para concentration is around 60% after 132 hours of operation at Lujan Center. This value is almost the same as that of natural conversion, but the value is not so correct due to uncertainty in the calculation results. We, however, would say that the *conversion occurred almost naturally*."

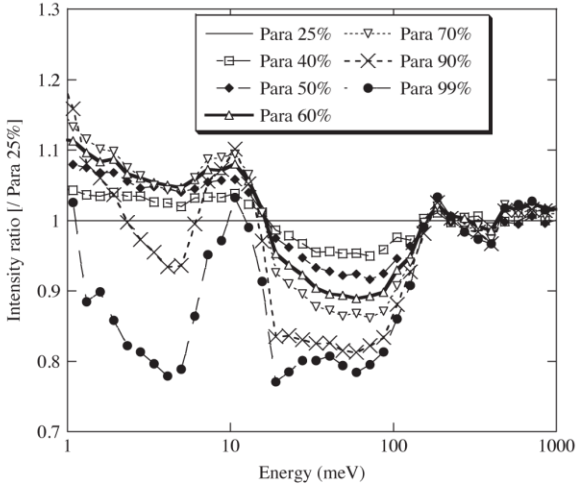


Fig. 3. Simulation calculation results ofi ntensity ratios at various ortho/ para ratios.

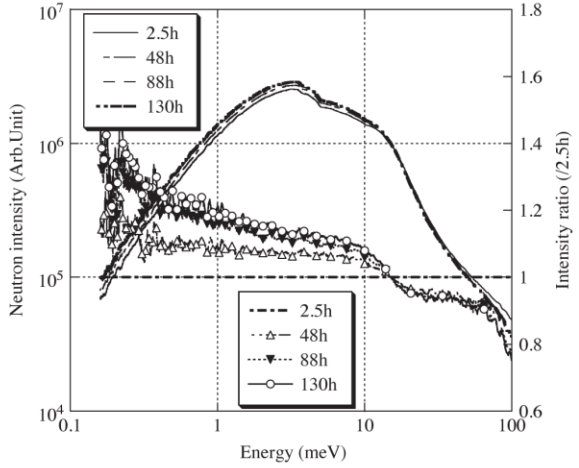
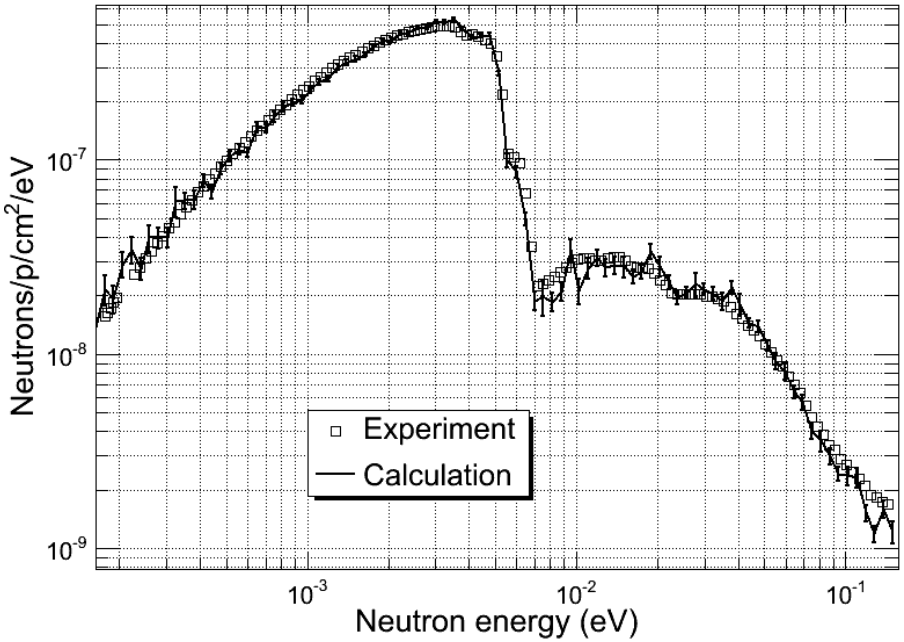
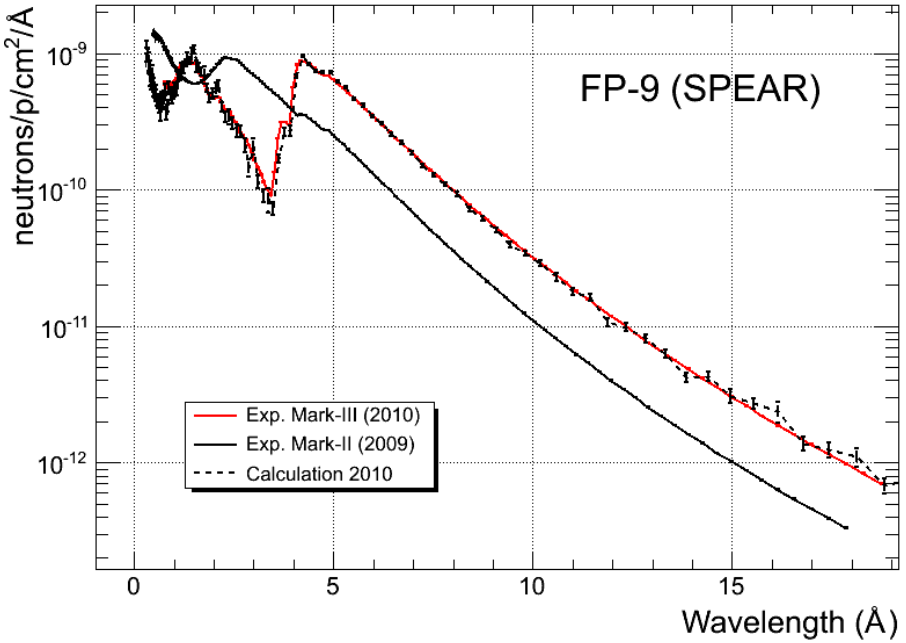


Fig. 4. Neutron energy spectra and intensity ratios at various hours after condensation.

Mark-III neutron spectrum



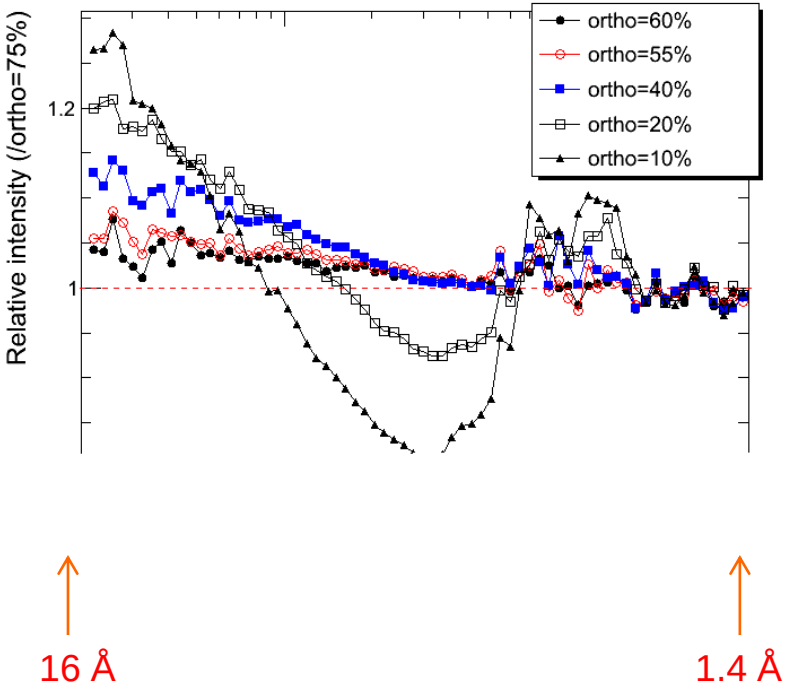


Calculation: O/P = 25%/75%

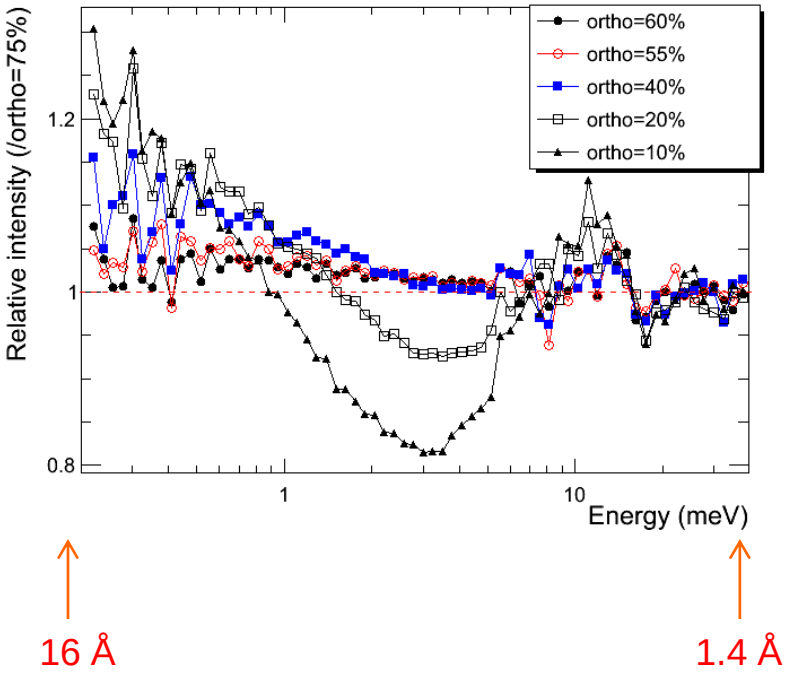
High Fidelity model including all details = collimators, slits, materials and fission detector

Calculated changes to the neutron energy spectra

Point detector tally (F5) at 10 meters with maximum field of view on emission surface of the moderator:

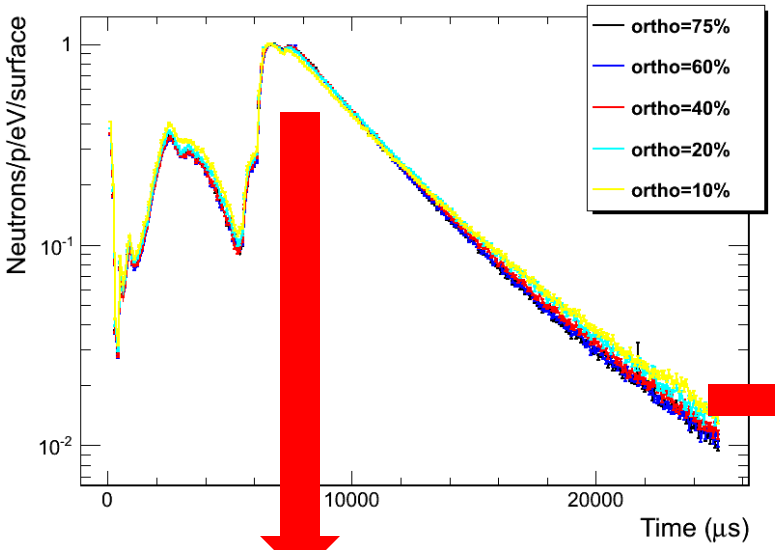


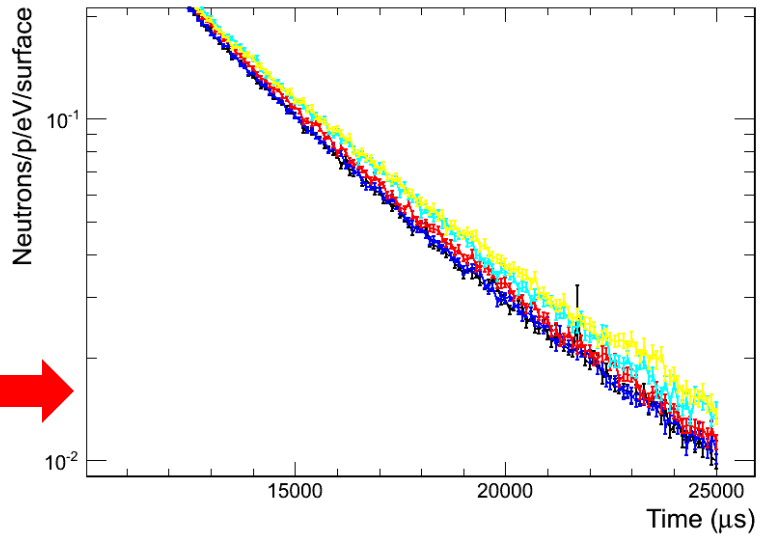
High-fidelity model of FP-9 collimation system, surface tally (F2) with U235 fission cross section folded:

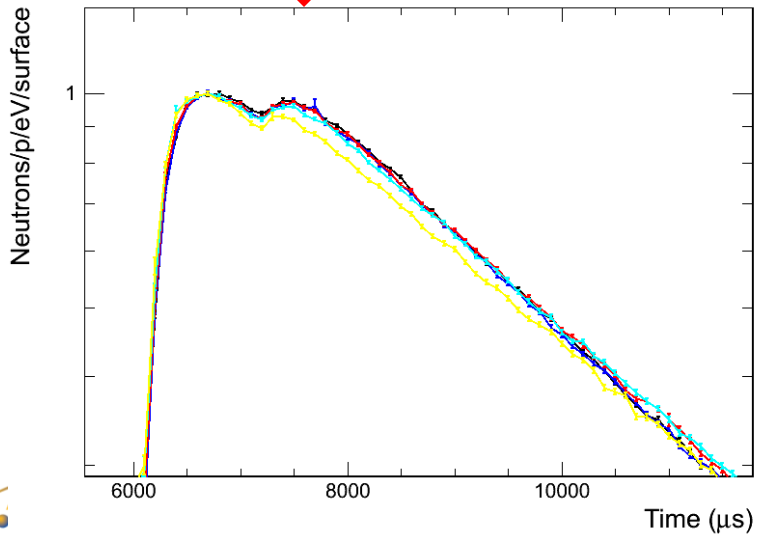


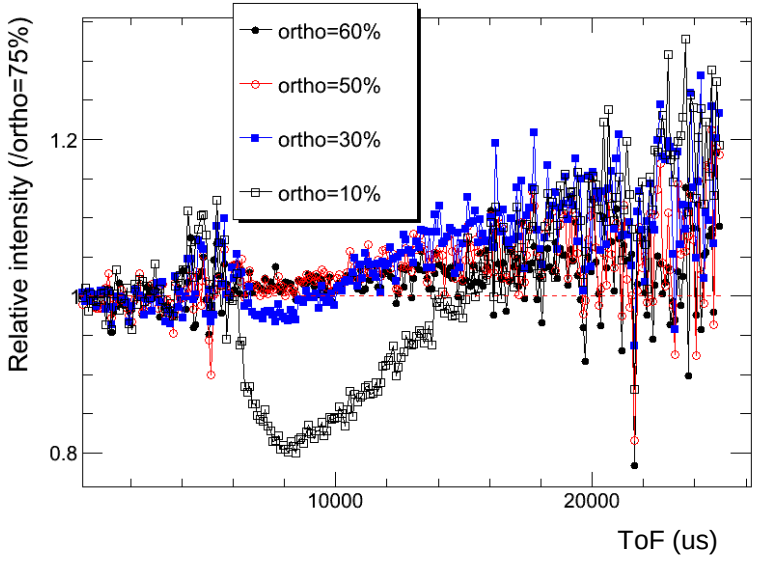
Calculations done using the new continuous representation scattering kernels.

ToF representation (surface tally)



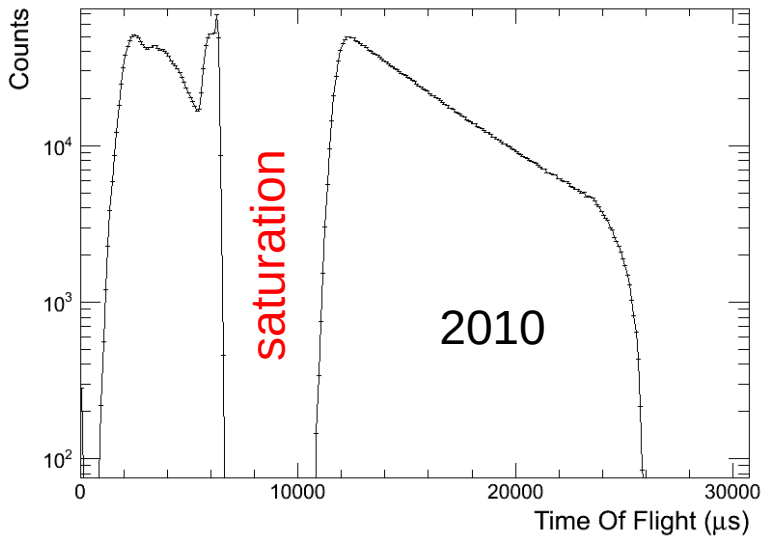


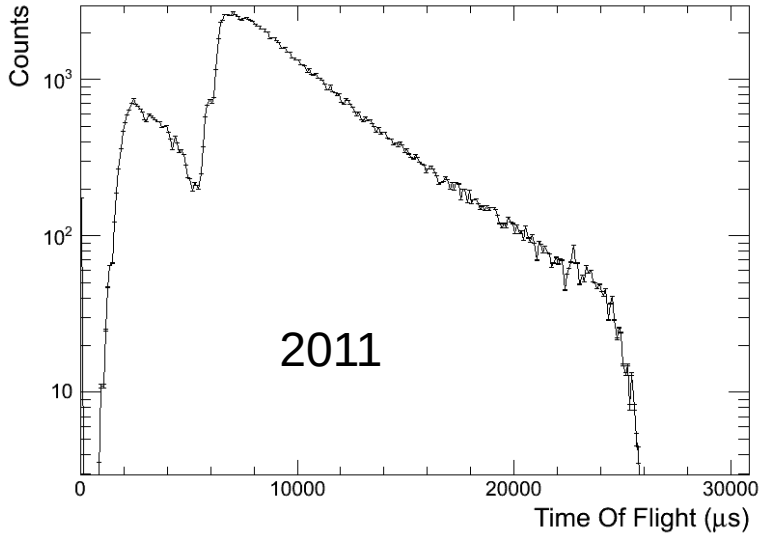




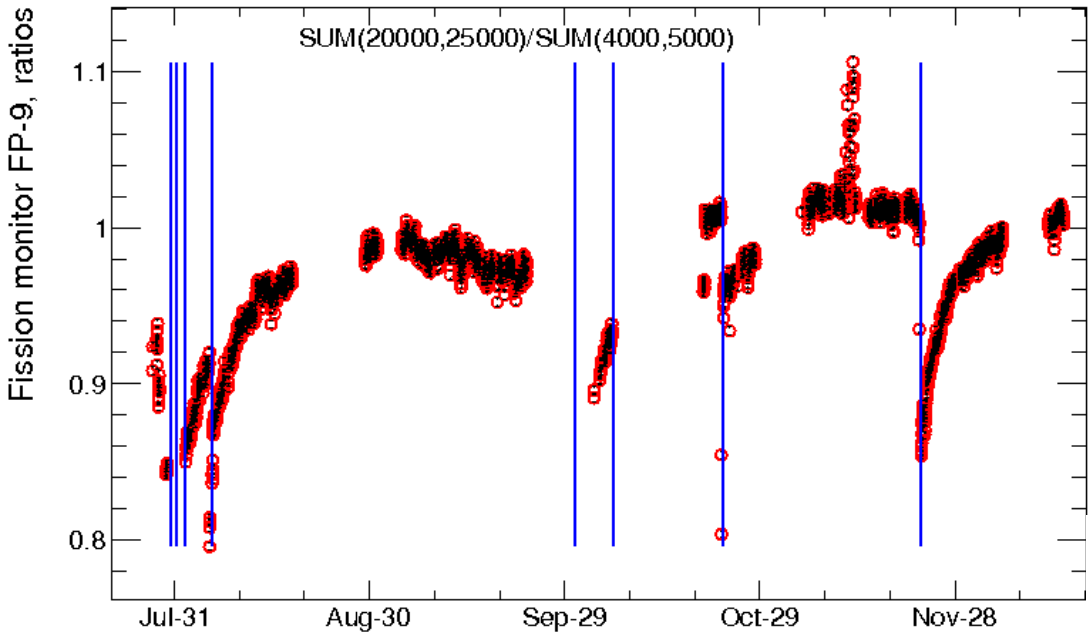
Neutron flux monitoring at FP-9

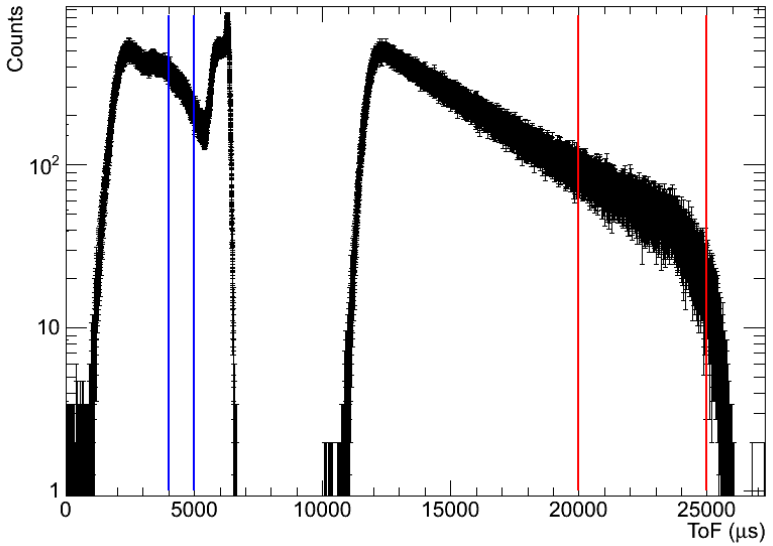
- **Low efficiency fission (U235) chamber (LND inc.)**
  - 2010: #3005 ($\varepsilon=10^{-5}$)
  - 2011: #3007 ($\varepsilon=10^{-7}$)
- **Installed upstream from the instrument (consistent beam conditions)**
- **Continuous data taking**
  - 2010: 10 min. runs
  - 2011: 15 min. runs



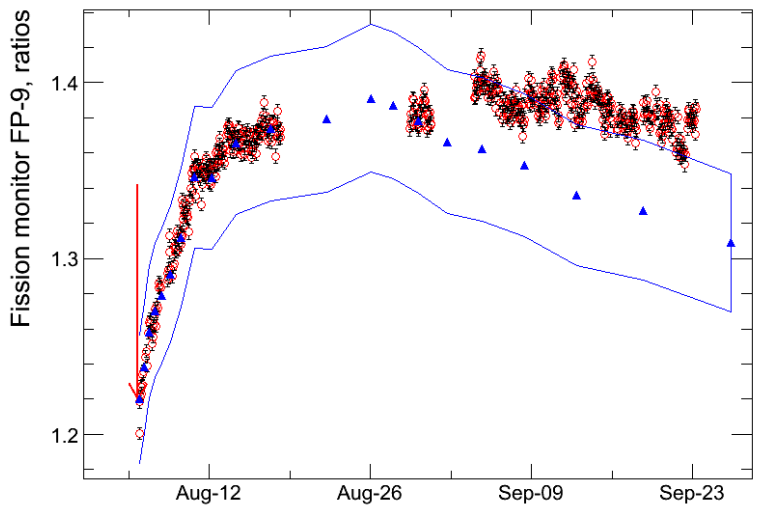


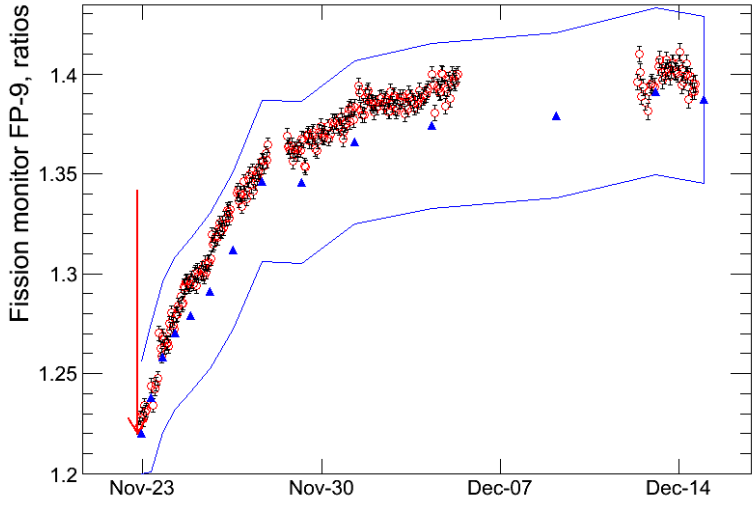
Run cycle 2010 measurements





Run cycle 2010 measurements cont'd

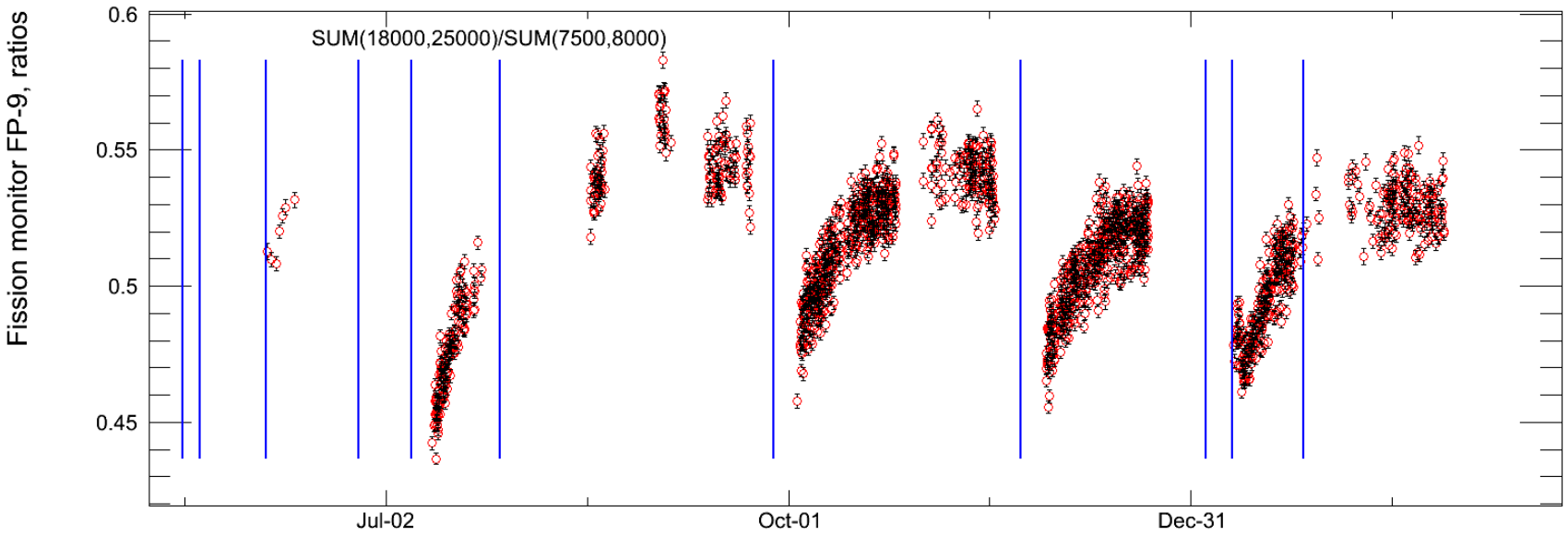


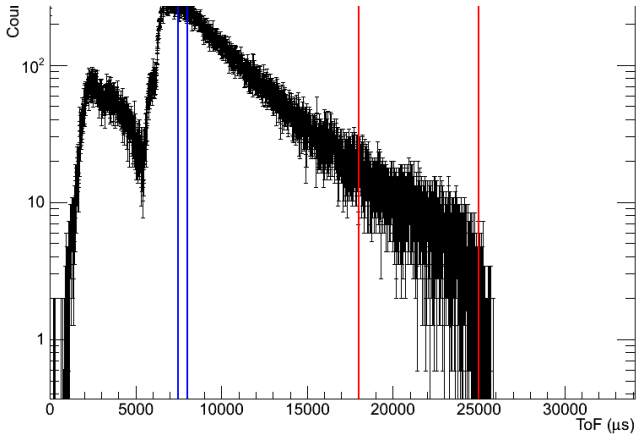


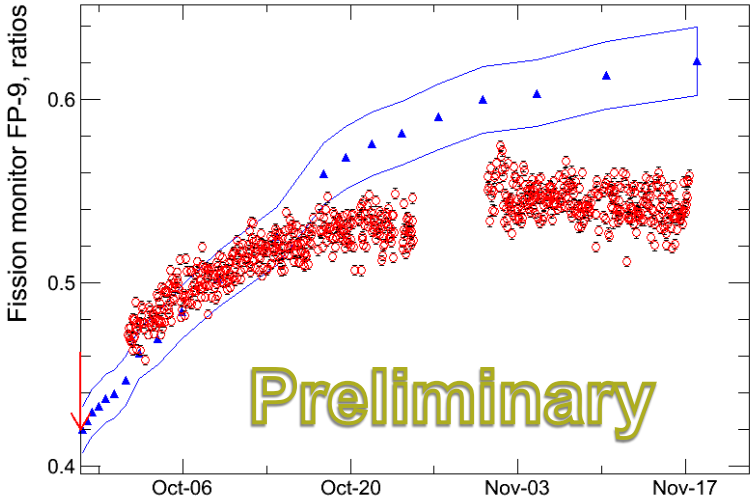
Experimental vs Calculated relative ratios

Ratio = (18000=>21000)/(2500=>3000)

Run cycle 2011 measurements







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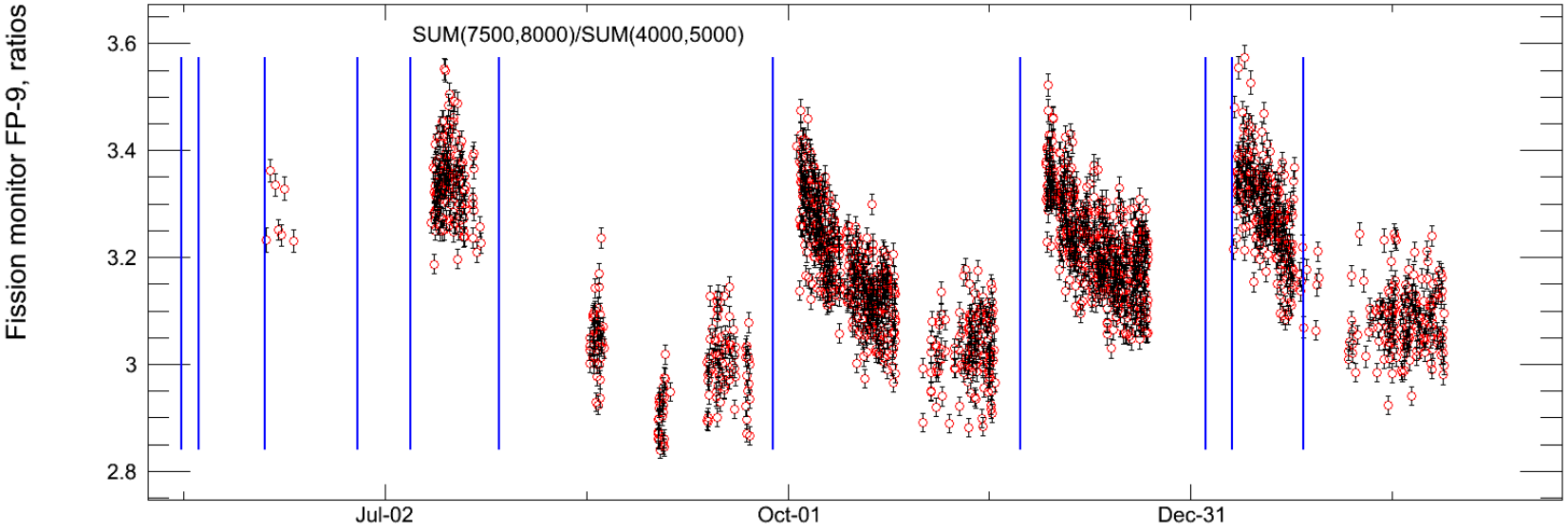
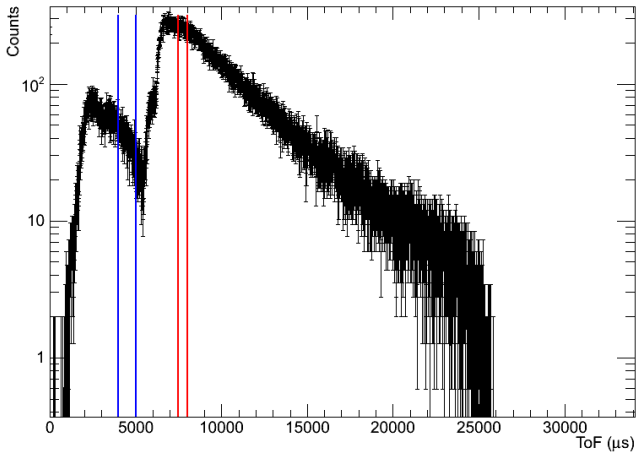
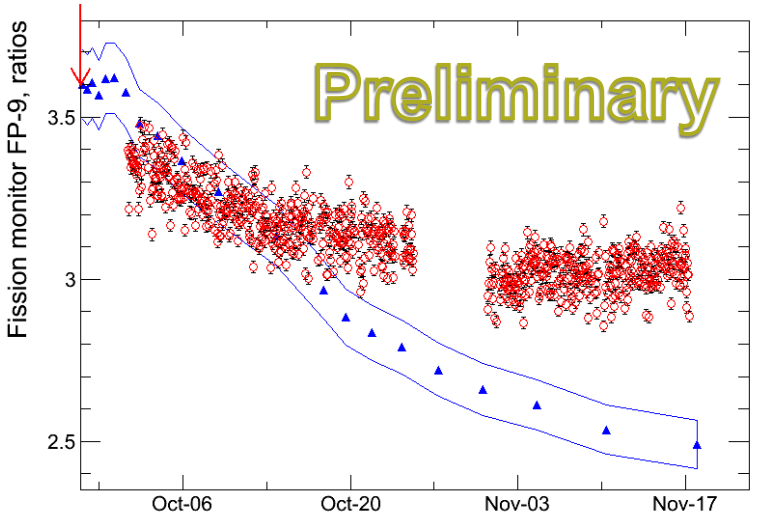
Run cycle 2011 measurements cont'd







Conclusions and future work

- **We setup a low efficiency neutron beam monitor at FP-9 at the Lujan Center**
- **Continuous data taking during 2010 and 2011 run cycles**
- **Data clearly show changes to the shape of the neutron energy spectrum over time**
- **The O/P conversion rate starts off similarly as that of natural conversion in 2010 data set**
- **The 2011 data set shows a deviation from natural conversion rate, inconclusive because of unknown starting O/P ratio**
- **We cannot determine the equilibrium point due to [relatively] short duration between vents (scheduled or unscheduled)**
- **Working on a capability to simultaneously measure the O/P ratio and the neutron production rates**