OAK RIDGE National Laboratory

A Real-Time Ortho-hydrogen Diagnostic for a Liquid Hydrogen Moderator

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Outline

- Ortho and para hydrogen are neutronically different, it matters, we want minimal ortho, we want to KNOW we have minimal ortho, and it's both complex and hard….
- The SNS PPU project, primarily meant to upgrade the accelerator complex to 1.3 GeV and 2.0 MW, also provided an orthohydrogen converter (catalyst) system and development for a real-time Raman based diagnostic to monitor the orthohydrogen level
- SNS FTS moderators were designed to be not particularly sensitive to orthohydrogen fraction, in part because we knew it would be difficult to ensure full equilibration during operation – so we really only needed to get FTS moderators down to about 1% orthofraction, which is still quite a bit off-equilibrium
- We tested an immersion probe diagnostic, which worked well enough to confirm that that radiation-induced backconversion prevented full relaxation of the hydrogen and would result in a significant power dependent variation in orthofraction during regular SNS operations
- We deployed a "through-the-window" diagnostic on each of the three hydrogen loops in the SNS CMS at the same time as we deployed IONEX based catalyst systems, and all appear to be working exactly as intended

Background

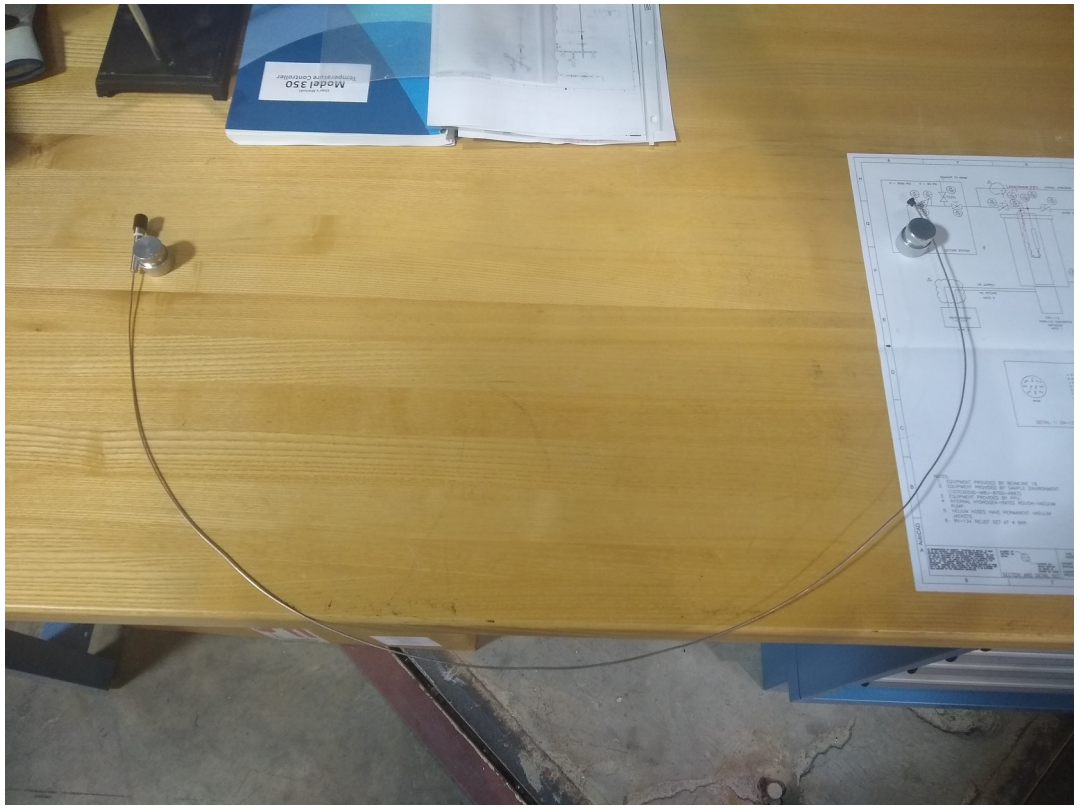
- Orthohydrogen and parahydrogen are different
  - spin coherence in parahydrogen reduces the cross section by >100x at 1 meV
  - equilibrium at 20 K is 0.2% ortho, but that represents 30% of the scattering cross section
  - an extra 0.2% changes the cross section by 30%; changing “thickness” from 28 mm to 38
- Relaxation is famously slow, taking months to get to 0.2% equilibrium (20K) without catalysts
- Kinetics model suggests that SNS power levels, around 0.02 W/cm^3, can result in a shift away from 0.2% ortho at equilibrium to as much as 30% ortho, on a bad timescale
- Add a catalyst – the system will settle quickly and to a near-equilibrium (low) ortho level which will let us better optimize the moderator performance at a higher level of performance
- But how will we know? Is it even needed? Is all this correct or naive conjecture?
  - ***Need a reliable diagnostic for real-time in situ measurements of orthohydrogen fraction in liquid hydrogen moderators under neutron irradiation***

Orthohydrogen Diagnostic Development Overview

- Measure ortho-para ratio using *in situ* Raman spectroscopy
  - Direct indication of ortho and para hydrogen levels in liquid flow
- Primary design: use cryogenic viewport to view circulating hydrogen with Raman head
  - Sapphire windows separating hydrogen volume, insulating vacuum, and optics
  - +Strong signal with lowest background - most sensitive measurement
  - +Maintain vacuum boundary to monitor for leaks
  - -Required demonstration of cryogenic window at pressure
  - -Needed physical system modification
- Backup design: fiber immersion probe penetrating circulating hydrogen
  - Sapphire fiber encased in steel with compression fitting
  - +Could deploy in CMS using existing penetrations without significant system modifications
  - -Weaker signal with higher background - less sensitive measurement

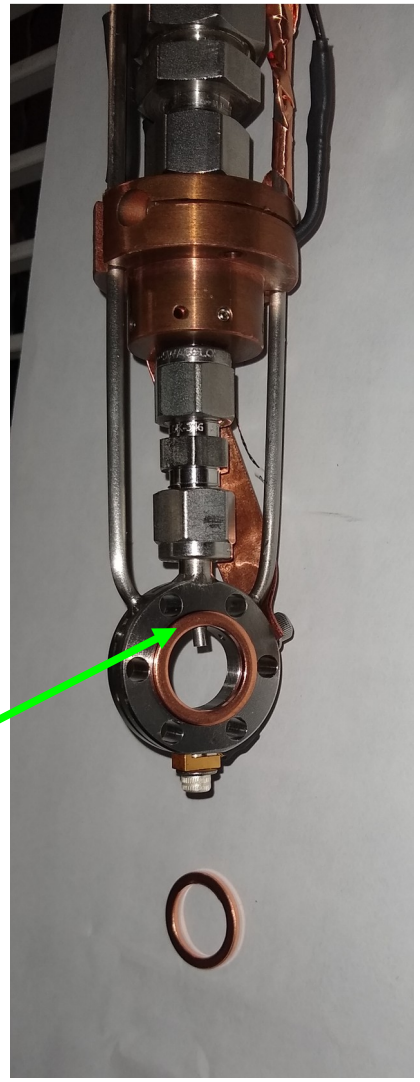
Probe Diagnostic
Commercial Probe, OTS Laser/Spectrometer

- 50” long, sapphire core, SS jacket, compression mounting
- Raman head mounted to fiber
- 50 mW Laser and spectrometer connected to head
- Fiber extends into liquid hydrogen volume replacing redundant thermocouple



Probe Diagnostic Testing in Bench-top Apparatus

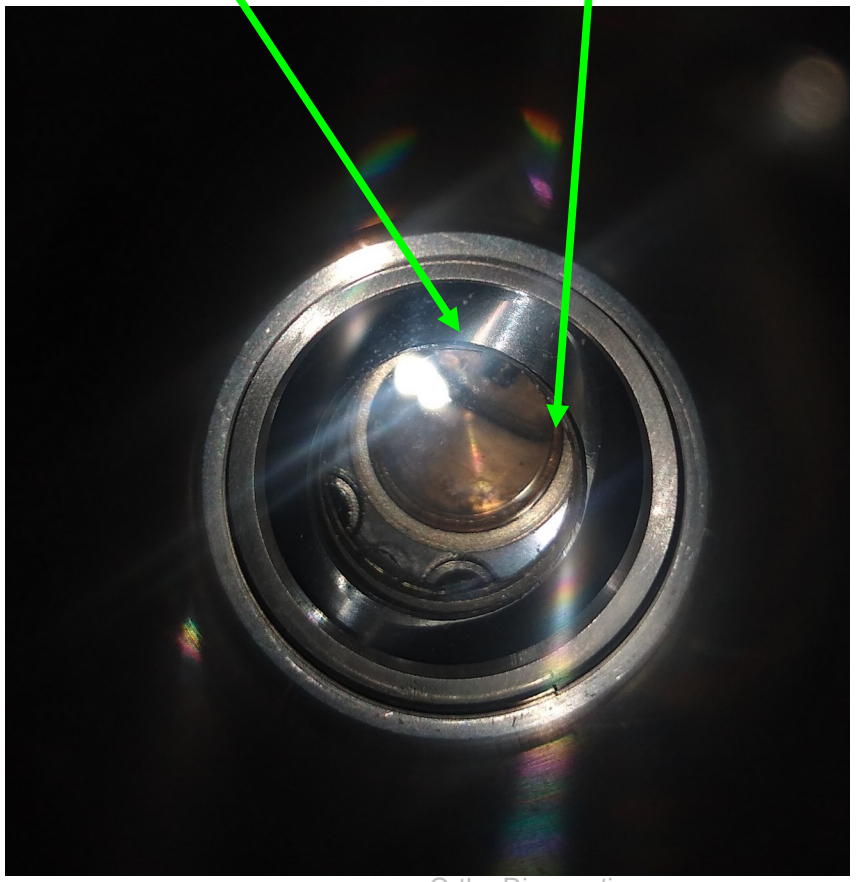
Mounted at test stand



Probe tip protrudes into sample cell

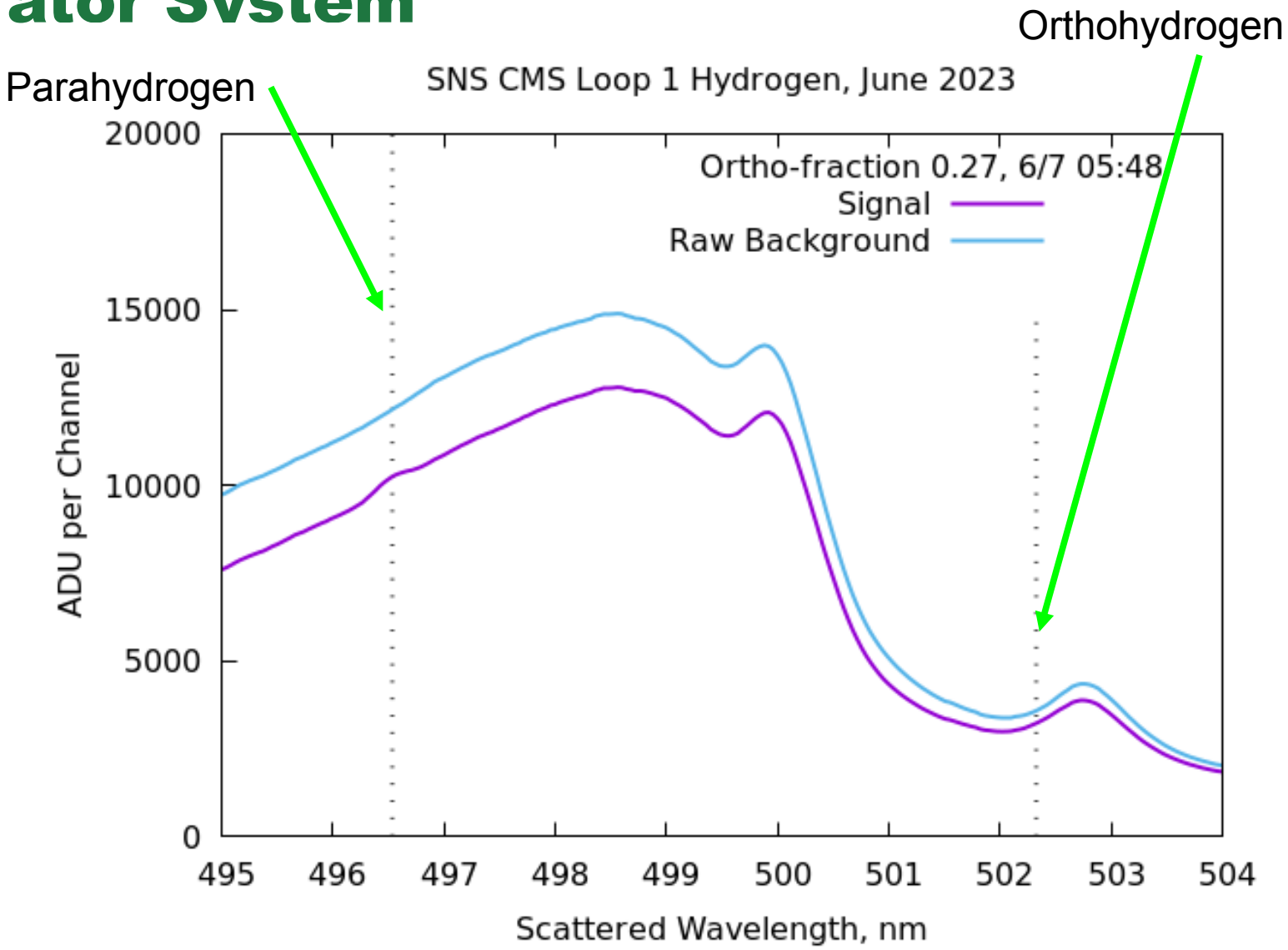
Fiber probe tip

Liquid hydrogen fill level



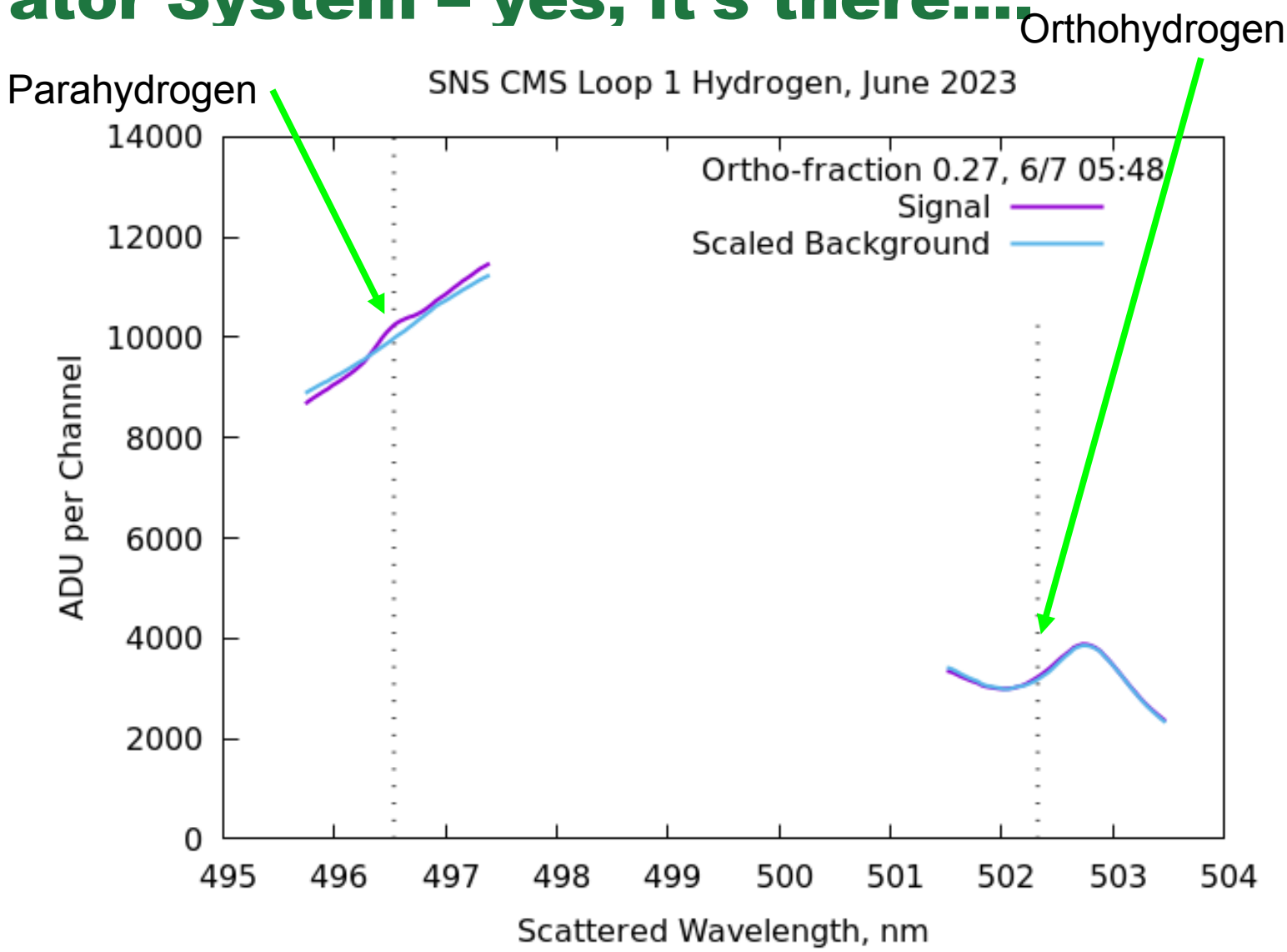
Probe Diagnostic Spectra in Cold Moderator Svstem

- Immersion probe tested on bench-top system was installed into existing CMS to test function and get a snapshot at orthohydrogen levels before installation of converter module.
- First attempt was in November 2022; ***fiber broke on insertion and yielded no data***
- Second attempt in May 2023 successful
- Background is high and varies over time



Probe Diagnostic Spectra in Cold Moderator System – yes, it's there…

- Careful treatment of data permits extraction of subtle signal
- 36 Hours of background measurements taken before hydrogen fill
- Best match background is scaled and subtracted from signal to reveal two peaks – one arising from parahydrogen transitions (0-2 state); one from orthohydrogen transitions (1-3)
- Peak area ratio gives ortho-para ratio



Probe Diagnostic Spectra in Cold Moderator System – no, REALLY

- Careful treatment of data permits extraction of subtle signal
- 36 Hours of background measurements taken before hydrogen fill
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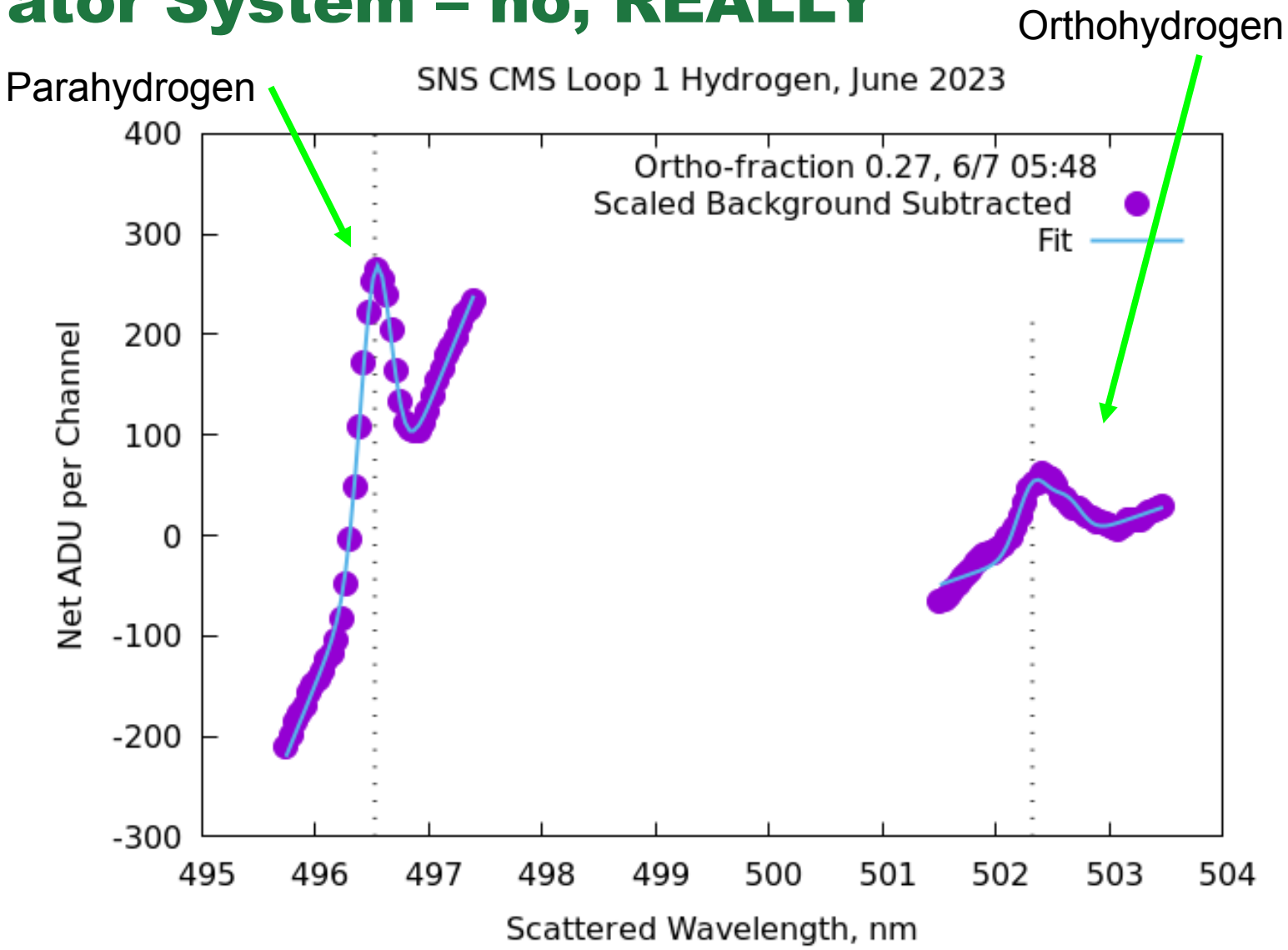
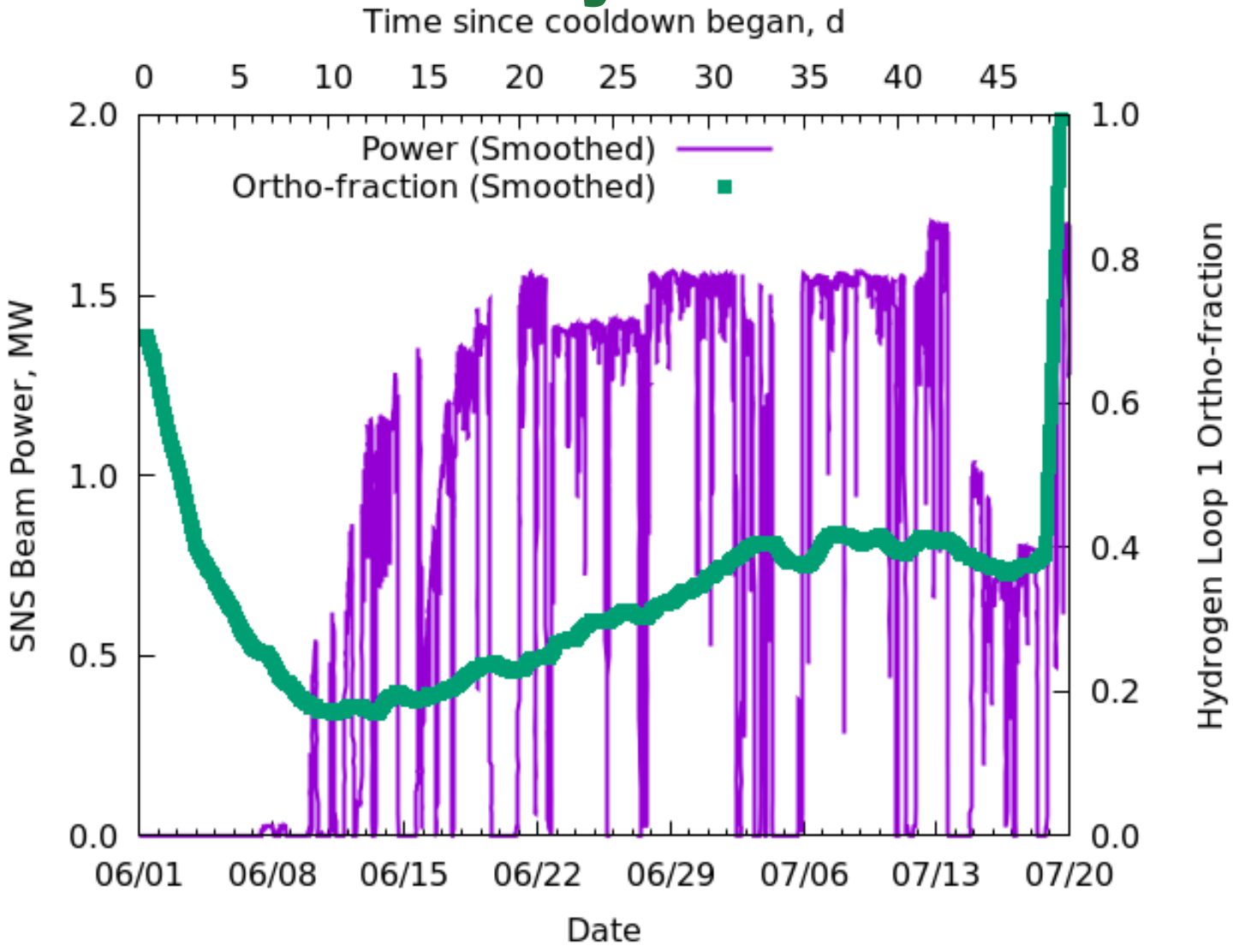


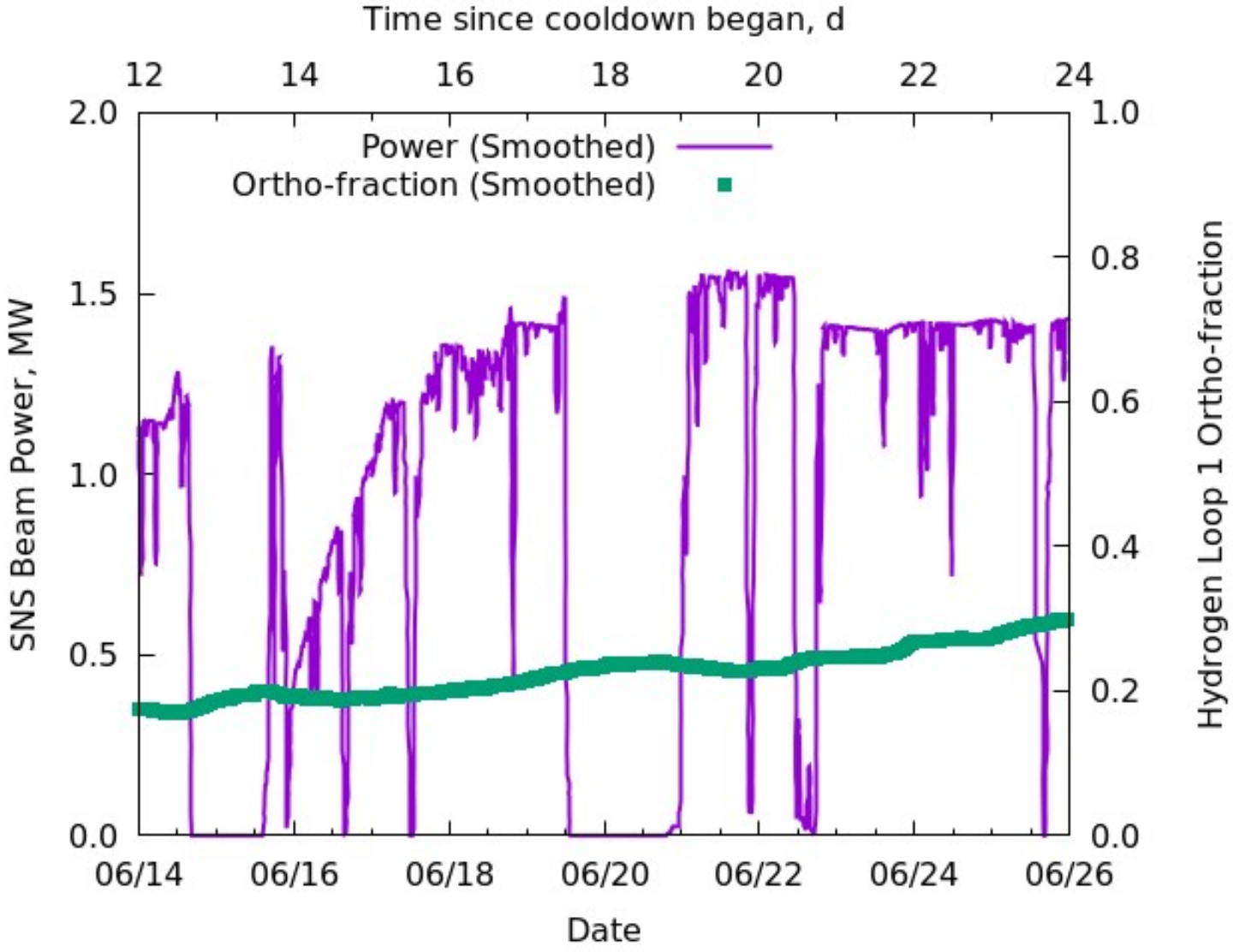
Probe Diagnostic Operational Results in Cold Moderator System

- 10 one-second spectra are averaged for display
- Every other spectra saved; 39 averaged together for analysis – one every 13 minutes
- 75% orthohydrogen hours after fill
- 27-30% after 5 days – consistent with natural relaxation
- As soon as beam comes on, orthohydrogen levels climb
- Steady state at 1.4-1.7 MW takes 4-5 weeks
- Kinetics model confirmed, diagnostic works at >2-5%



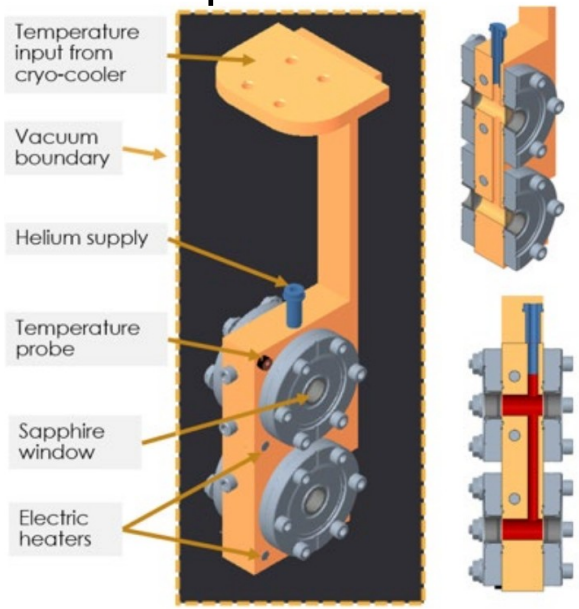
Probe Diagnostic Operational Results from Power Transient

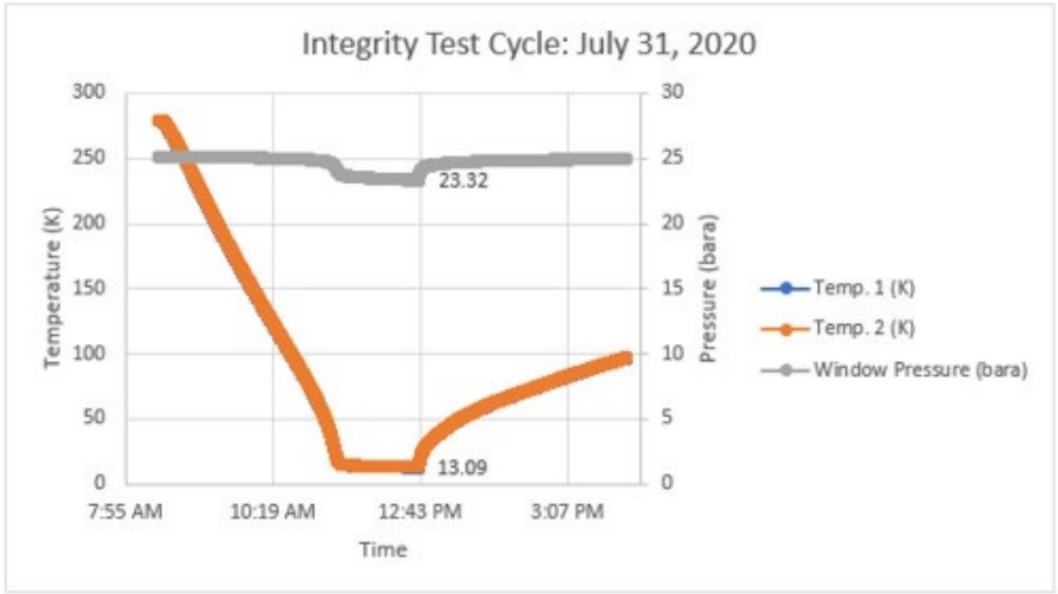
- Orthohydrogen levels respond to beam off periods with lag
- Early beam-off periods show continued increase in orthohydrogen levels for around one day before beginning to fall again
- Later beam-off periods (once some sort of equilibrium has been achieved?) show faster drop
- If we weren't adding a catalyst, we should not fill so early



External Window Diagnostic
Window Requirements and Testing

- Commercial sapphire windows are available – certified for temperature OR pressure, not both
- Needed to verify that windows would not fail at operating condition even after multiple cycles
- 20 K, 19 bar relief / 14 bar operations, 3-4 cycles per year expected conditions
- 13 K, 24 bar, 30 cycles needed to demonstrate fitness, 1 day needed per cycle
- Tests performed at ORNL's Pellet Laboratory – success; no leaks after 30 cycles / 4 windows



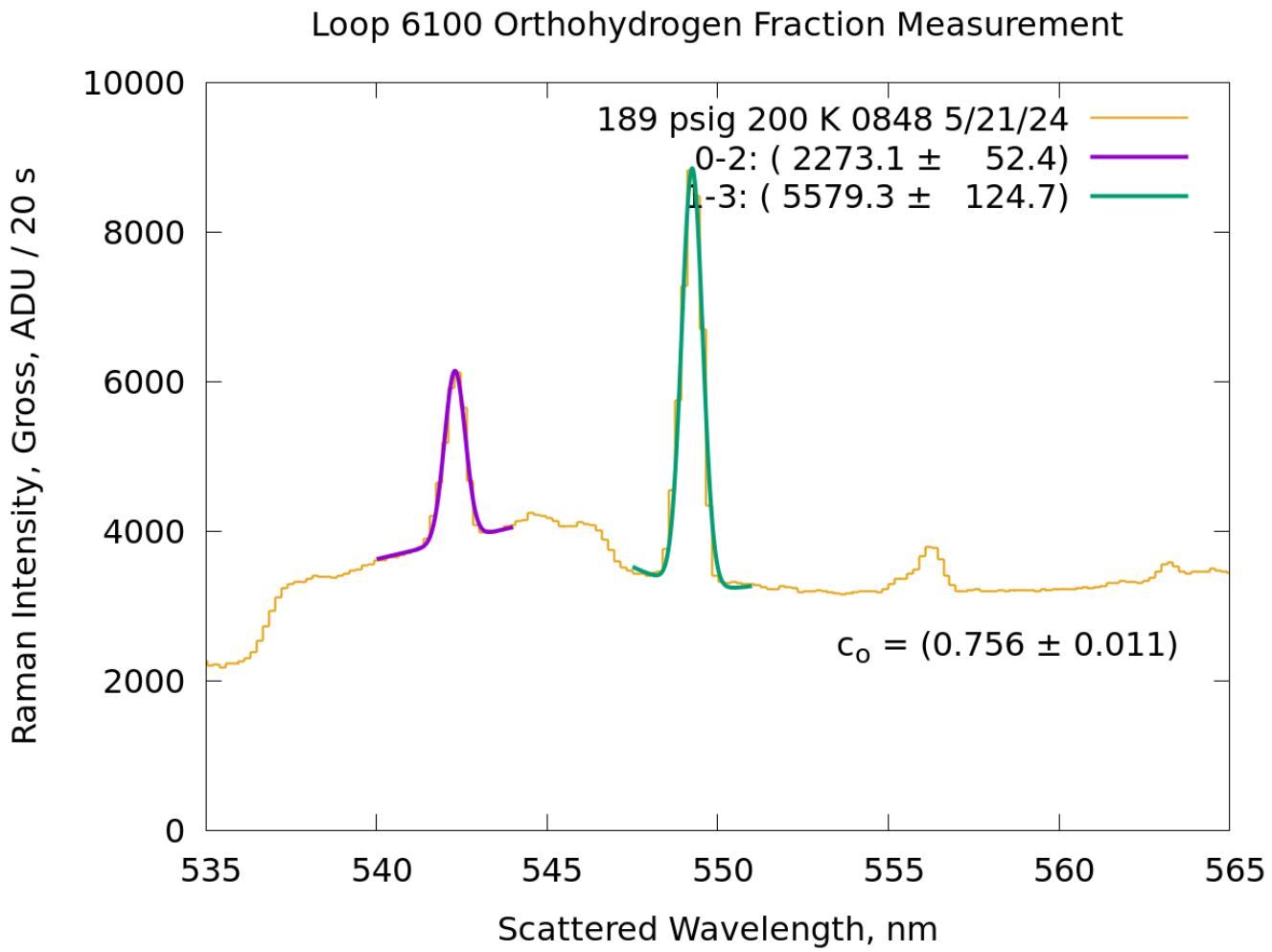


External Window Diagnostic Raman System Installation

- Windows are welded into 100 mm dead legs in the catalyst system, adjacent to external windows
- Raman heads with 100 mm working distance probes look into the windows
- Each Raman head is coupled to an optical spectrometer, all read by a single computer
- Each Raman head is fed by an independent laser (100 mW at 532 nm)

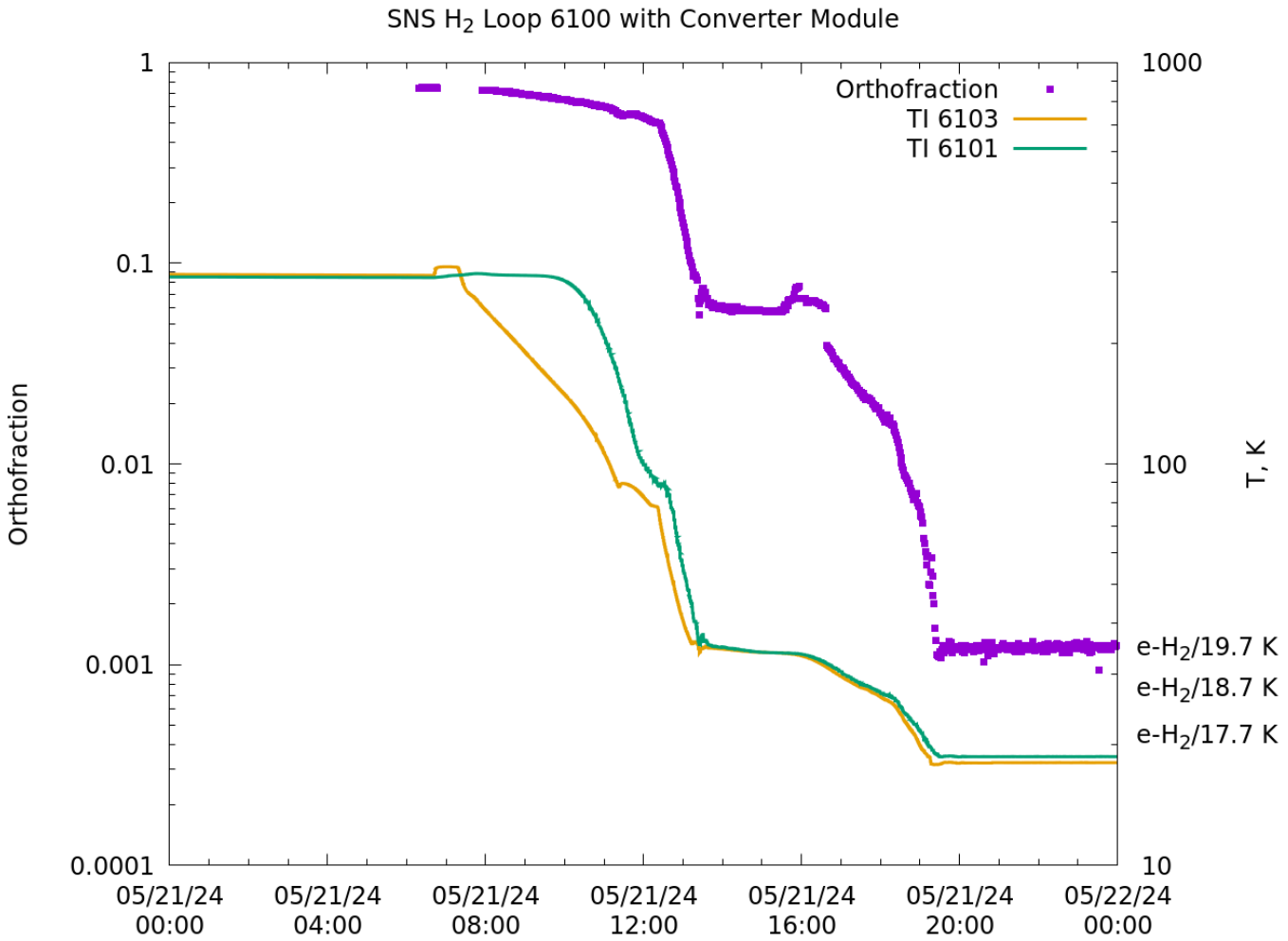
External Window Diagnostic Room Temperature Spectrum

- Recall that very first Raman spectra from the probe installation, where there was significant data processing to see, let alone quantify, the spectrum
- When we replace 1200 mm of sapphire with 3 mm of sapphire, the background goes way down and the signal goes way up
- Instead of 15 minutes worth of data collection, we need 20 seconds and the peaks are obvious
- Plot shows raw spectra, without even dark current subtraction
- 300 K, 14 bar, < 30% of operating density



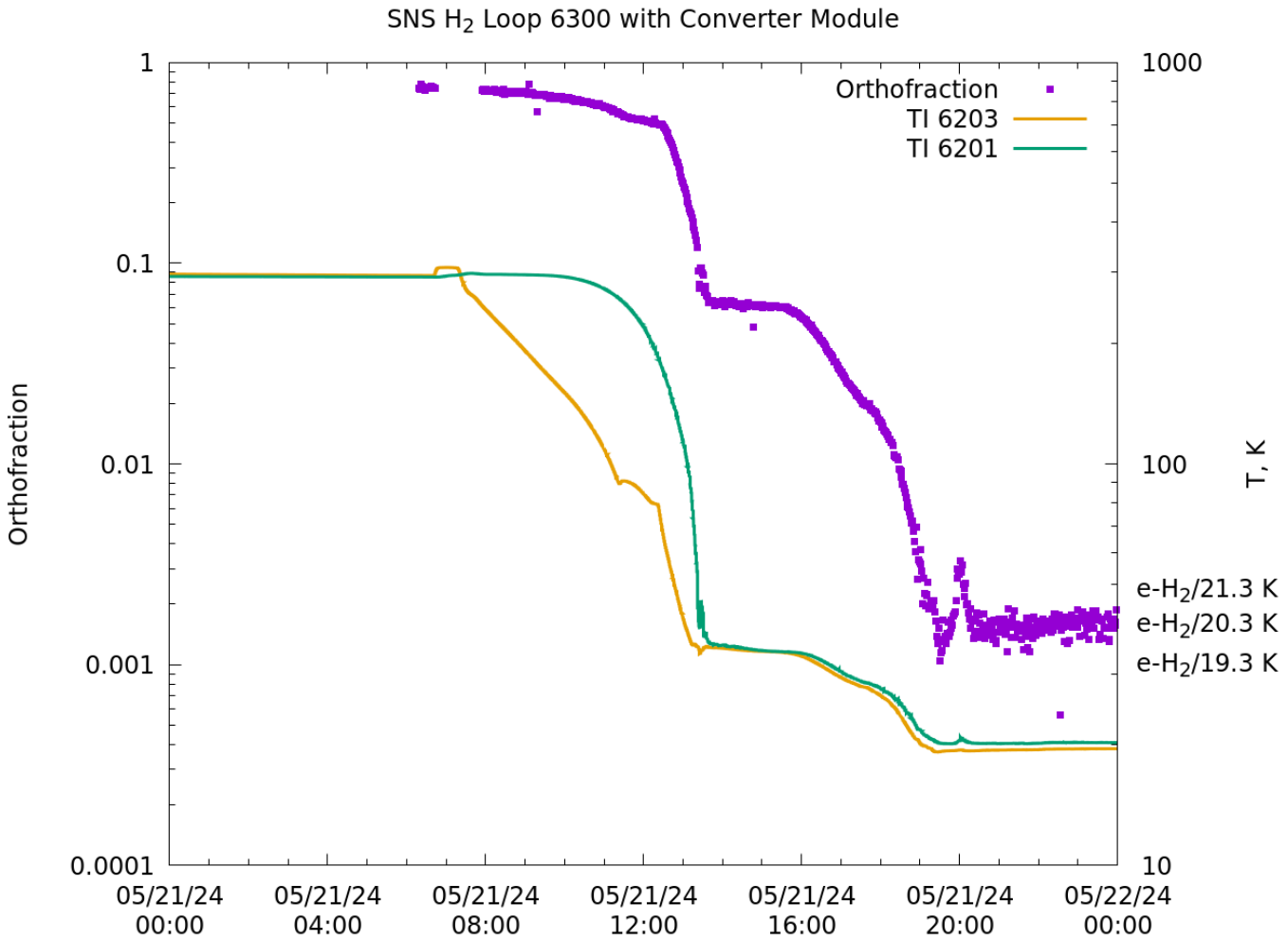
External Window Diagnostic
Calibrate at Room Temperature and Go

- Standard filling procedure gives us sufficient time at room temperature to calibrate conversion factor for ratio of relevant peaks to orthofraction
- First try, everything “just worked”
- Loop 1, Top Upstream Decoupled Hydrogen
- Diagnostic indicates that orthofraction falls to equilibrium values corresponding to a temperature within 1 K of thermocouple readout, which is better than we can really trust such a thermocouple
- Diagnostic indicates that orthofraction follows temperature within seconds, not months, suggesting that catalyst works well



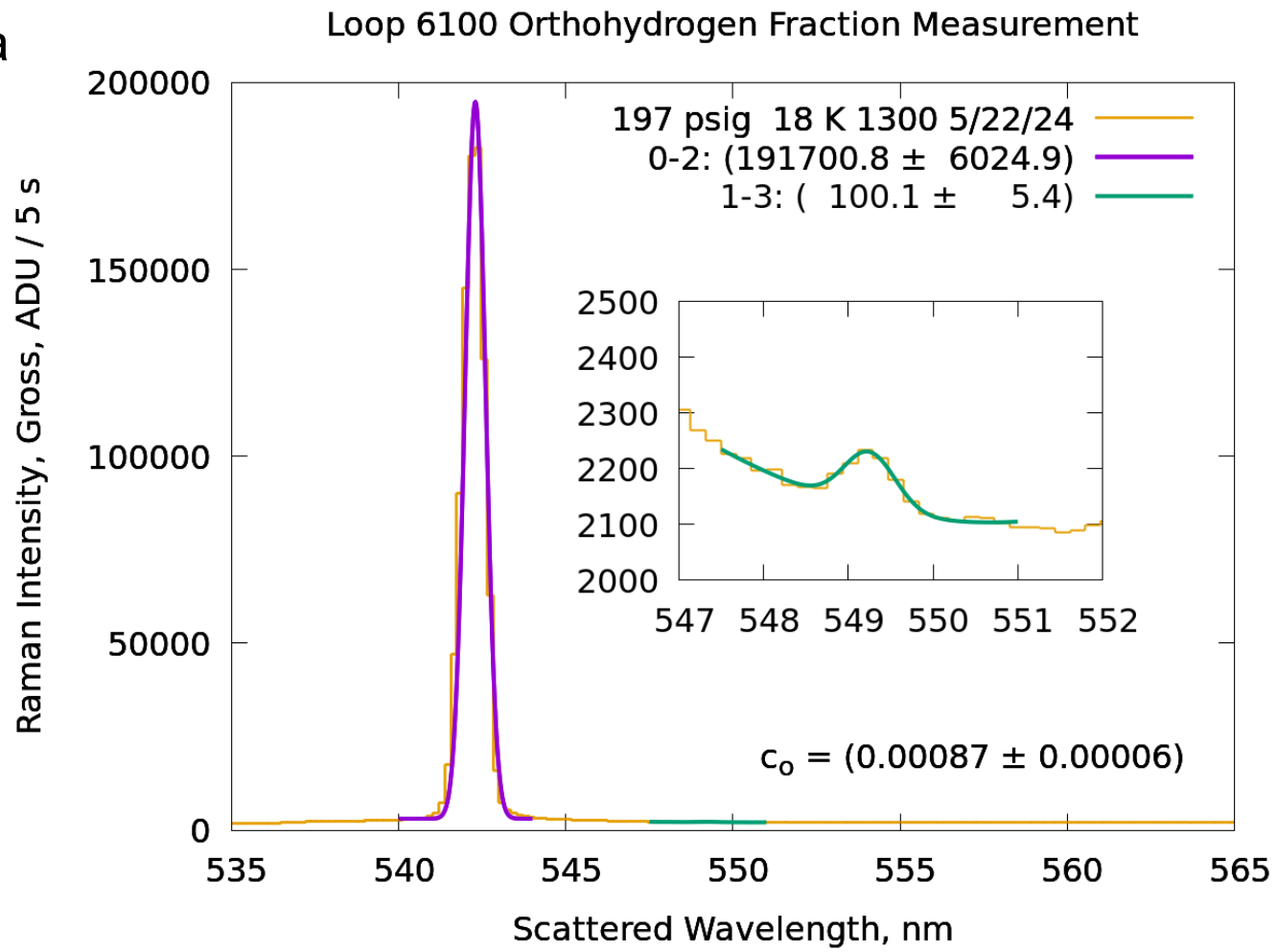
External Window Diagnostic Comparable Results for Different Loop

- Loop 3 (bottom downstream coupled moderator) goes to a slightly different temperature
  - Diagnostic sees that
- Small temperature hiccup after equilibration
  - Diagnostic sees that
- Still has a negligible response time
- Spectra are recorded here for 1 second, limited by SIGNAL, not background
- 30 spectra averaged for general periods
- 5 spectra averaged for closer looks



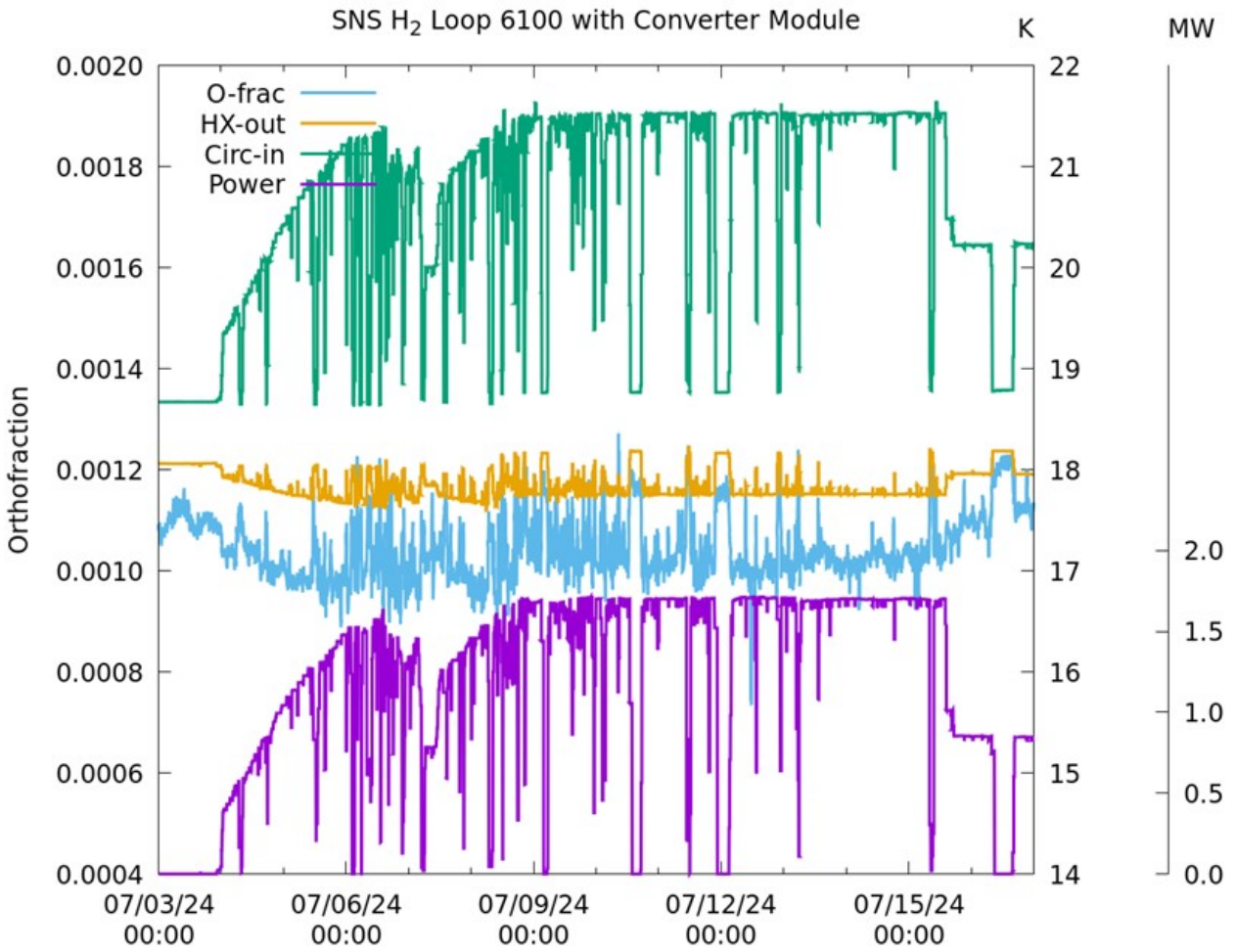
External Window Diagnostic Challenging Dynamic Range

- At room temperature, when the ortho:para ratio is 3:1, peaks are relatively comparable
- At operating temperature, para peak is dominant (~2000x) limiting collection time
- Ortho peak is still sufficient to see small changes in temperature
- May try filtering para peak to improve sensitivity even further, but may not be needed



External Window Diagnostic Challenging Dynamic Range

- Minimal response of orthofraction to power transients
- Temperature at HX output temperature is adjusted by heater (on the helium side) to offset power changes
- Power goes off, HX output temperature goes up by 0.2 degrees, orthofraction goes from 0.0011 to 0.0012
- Over 12+ days, going from no power to 1.6 MW, the orthofraction never changed outside of 0.0009 – 0.0012 (the catalyst works just fine), and we can reliably see differences of 0.0001 (the diagnostic works just fine)



Summary

- The “through the window” Raman probe works as designed – it can measure the orthohydrogen fraction in a working liquid hydrogen system every few seconds, with straightforward calibration, down to 0.001, with precision of around 0.0001.
- The immersion probe system does work – it can measure the orthohydrogen fraction down to some 0.01 to 0.02, which just isn’t good enough for our particular application
- The catalyst system deployed is more than sufficient to handle backconversion, and to reduce transient times to seconds
- The irradiated hydrogen kinetics model first presented at the Neuss ICANS is largely correct
- Resulting SNS hydrogen moderators are reliably stable, and in some ways feature improved absolute performance as well
- The orthohydrogen question has been a long-term effort from many different colleagues at many different facilities, and has been the subject of at least 30 ICANS papers since 2000.

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