Industrial development of large dichoric filters and optimization of substrates using atomic layer deposition.

Abstract: ***I will report on recent completion of a joint R&D project between RAYTUM inc. and BNL on development of optical coatings on large area substrates appropriate for high energy physics applications; particularly for liquid argon scintillation detection.***

Topics:

1) **some background on dichroic interference filters**
2) **Technique of Atomic Layer Deposition and contrast with PVD**
3) **Performance on uniformity**
4) **Status of R&D for production**

6) **Other R&D facilities at BNL**

Milind Diwan Sep 10, 2024

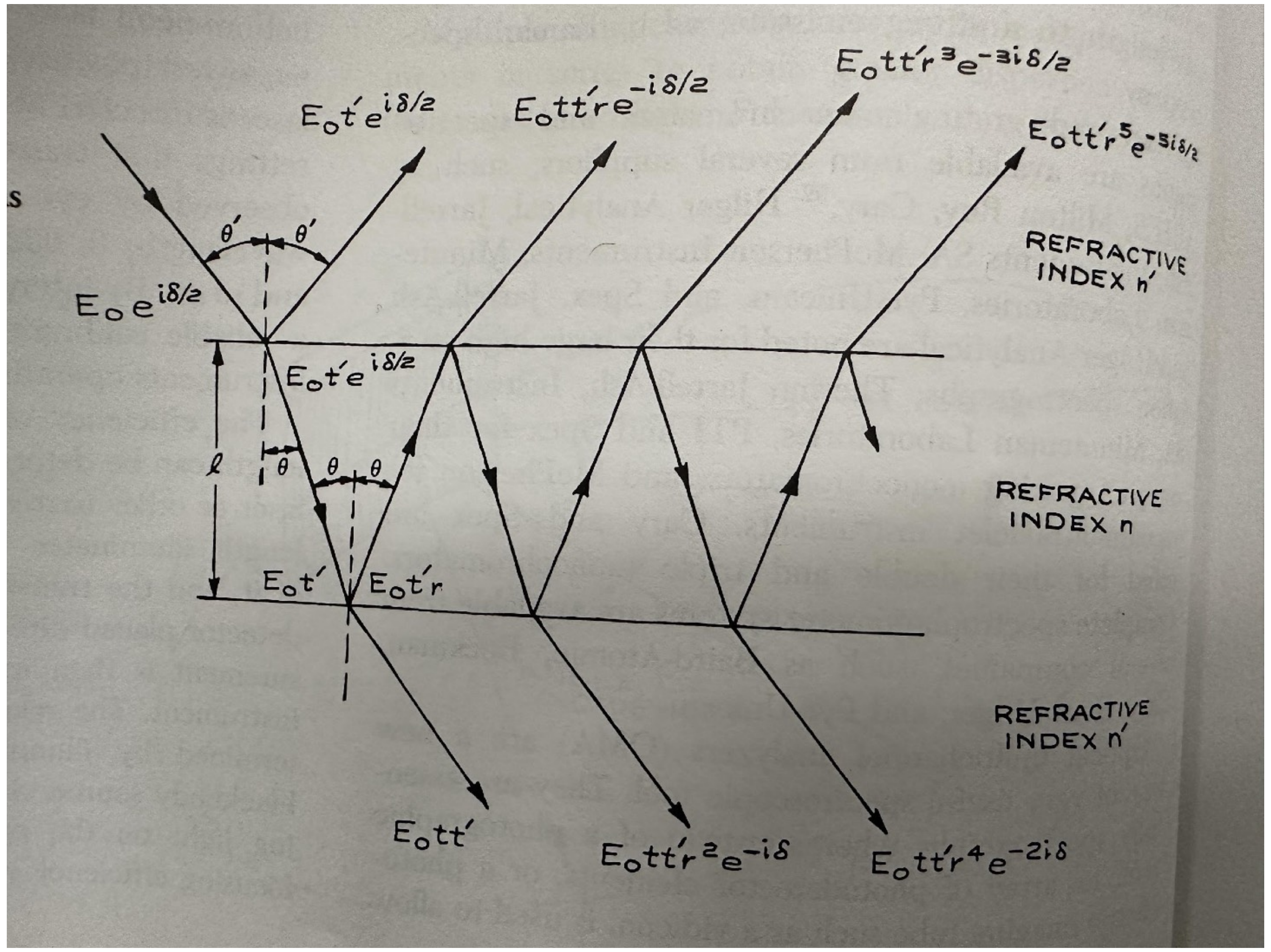
Thanks to RAYTUM Photonics

Summary

- Progress
    - The SBIR project will complete in September 2024
    - The project has been successful at designing the producing prototype filters. Large (DUNE 1 size) Filters on substrates of Saffire and B33 glass have been delivered.
    - Performance of these has been measured.
    - The process has been optimized for producing very large filters (up to 50 cm x 50 cm).
    - Coatings of wavelength shifters can be placed on one side of the filters. If requested this work can be done at modest cost.
    - BNL also has the capability for PTP and TPB coatings, but some investment will be needed.
        - New photo-spectrometer is being built to make absolute measurements in the lab.

Reminder of interference filters

Basics of an etalon or two parallel mirrors.



Moore, Coplan, Davis

- If phase shift and absorption is neglected on interfaces.
- $\frac{I_T}{I_0} = \frac{1}{(1 + \frac{4R}{(1-R^2)} \sin^2 \delta/2)}$ where R is the reflectivity and $\delta = 2kl\cos\theta$
- Transmission maxima happen when
- $l = \frac{m\lambda}{2\cos\theta}$
- For a fixed thickness, max wavelength will move lower with angle.
- There will be transmission peaks at l/m values.
- A stack of etalons can be modeled by software to create a bandpass filter. Each etalon is made of a high quality dielectric layer with different index of refraction.

How does one deposit the layers ?

There are many techniques; but the state of the art is consider atomic layer deposition.

Unique Advantages of ALD vs Physical vapor deposition(PVD)

1. Precise and easy thickness control in a monolayer scale over established PVDs for high performance optical filter fabrication
2. Excellent uniformity (<0.1-1%) for fabricating large area optical components.(up to sub-meters)
3. Super conformity on Non-planar optical surface and components.
4. Low temperature process on plastics by energetic ALD.
5. Continuous and pin hole free.
6. Diversity of materials for DF designs.
7. Low maintenance cost and high volume manufacture is established.

https://www.veeco.com/products/savannah-thermal-ald-for-rd/

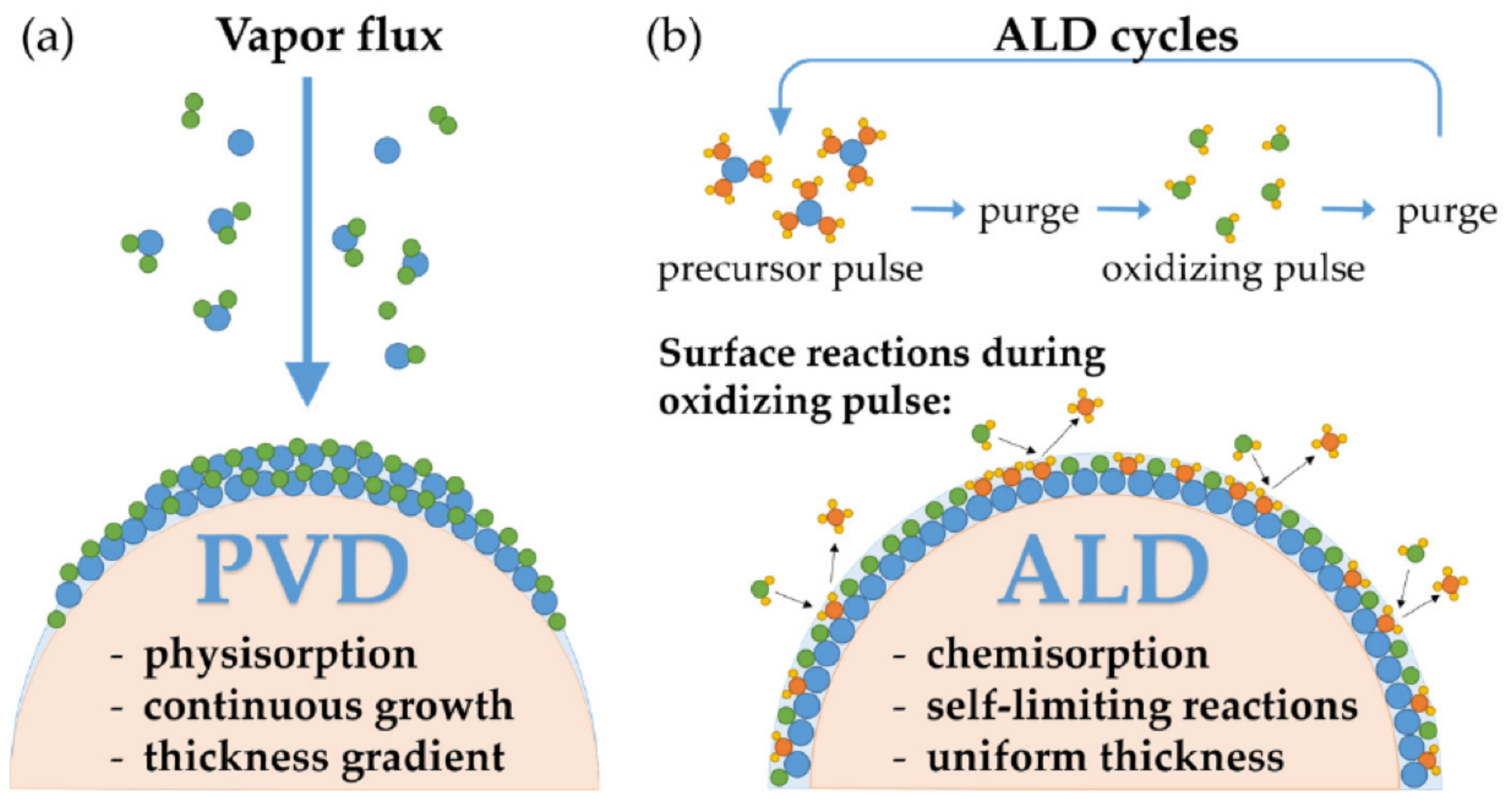
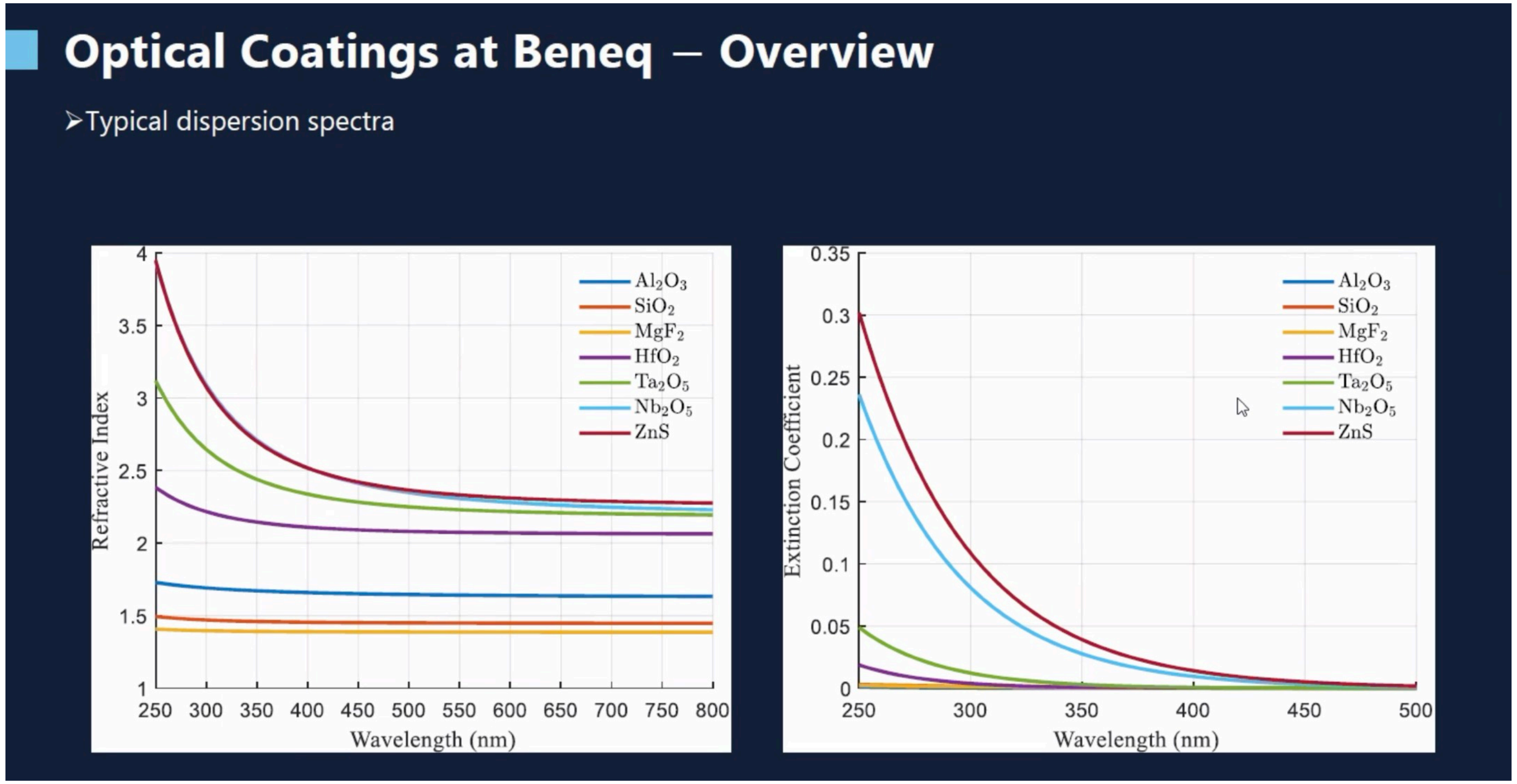


Figure 1. Illustration of (**a**) physical vapor deposition (PVD) deposition and (**b**) atomic layer deposition (ALD) on a hemispherical lens.

Key technical issues

Coating materials (high and low index), Substrate and stresses.

Materials	*Coefficient of line expansion(×10-6/°C at RT)*	*Refractive index at 500nm*
TiO2	*9.19(//C); 7.14(^C)*	*2.54*
Al2O3(sapphire)	*6.7((//C); 5.0(^C)*	*1,77*
SiO2(fuse silica)	*0,55*	*1,46*
Si	*4,2*	
N-BK7	*7,1*	*1,52*
Borofloat (borosilicate from SCHOTT)	*3,25*	*1,52*
Soda lime glass	*8,1*	*1,528*



- *There are other optical materials also, but practical list is limited.*
- *Substrate has to be chosen so that the CTE is reasonably matched to both coatings.*
- *The problem with fused silica is the mismatch of CTE. But this is getting resolved.*

R&D project timeline and scope

BNL and Raytum collaboration

- R&D project was proposed in 2021 with Understanding the scientific requirements.
    - A miniworkshop was organzied with both liquid argon and water based liquid scintillator experts Oct 17, 2022
  - Development of test samples from Raytum adapting ALD technology.
  - Testing of samples in real conditions for
    - Material compatibility, Handling issues, Detector performance
  - Understanding requirements for large scale production.
  - Preliminary development of production techniques.

The entire scope has been accomplished. However, it would be best to use the produced filters in a real detector setup.

Design/ variations/ and production technique are all in hand

Technical Challenges

1. Down-select ALD materials (low index(L), high index(H) with optical properties like n, k, optical loss as well as mechanical properties comparable with or better than well-qualified optical coating material by PVD.
2. Demonstrate a single layer ALD process with excellent uniformity on larger area using our innovative step rotation mechanism.
3. Fabricate multilayer filters based on our design with precise thickness monitoring and stress management.
4. Evaluate small optical absorption and scattering loss and filter performance setup and modelling refinement and process optimization.
5. Identify or modify an ALD tool from R&D for future volume production at low cost, such as spatial ALD or larger batch process ALD.

Updated Specifications for variety of projects

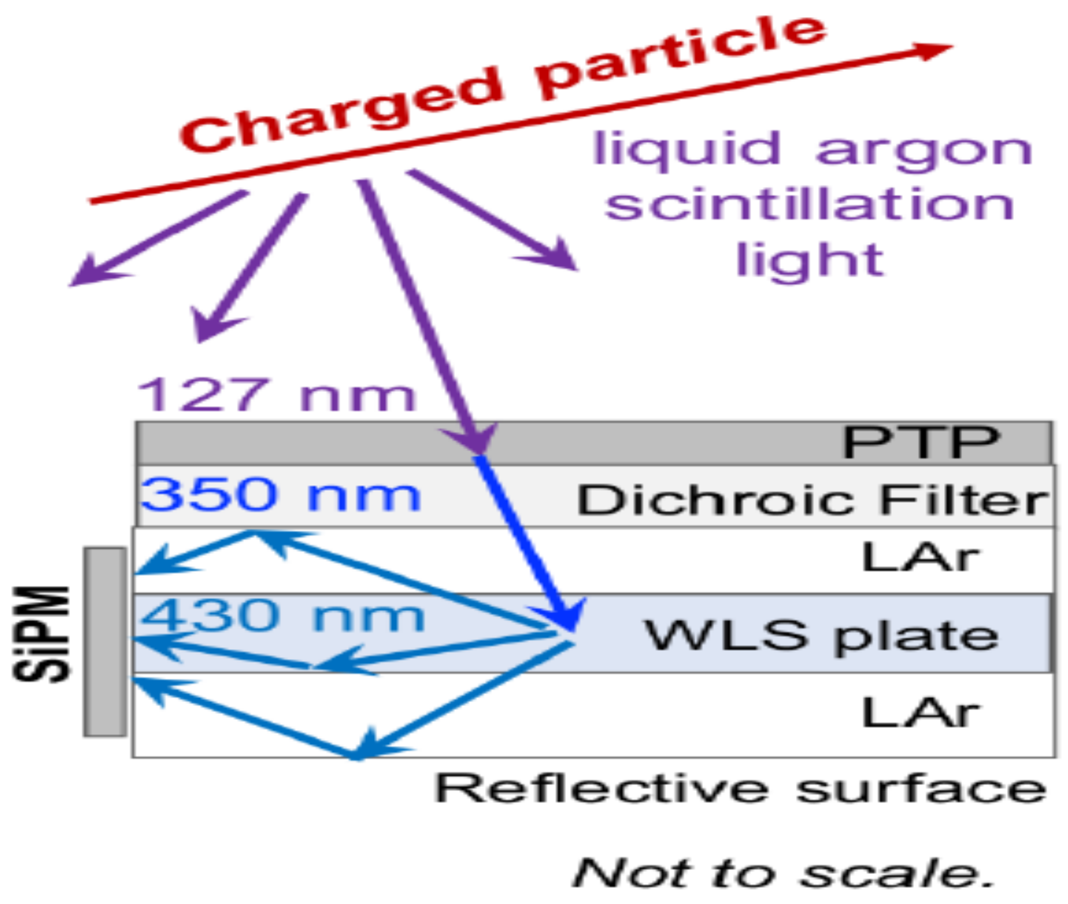
	DUNE Module 1	DUNE Module 2	DUNE Module 3 or 4	THEIA
Status	Technical Design Ready (ready for construction)	Technical Design Ready	Pre-concpetual	Pre-conceptual
Size	10000 tons	10000 tons	10000 tons	25000 to 100000 tons
Technique	Liquid Argon Scintillation	Liquid Argon Scintillation	Liquid Argon Scintillation	Water Cherenkov and Water Based Liquid Scintillator
Type of filter needed	Low pass	Low pass	Low pass	Both low and high pass
cut wavelength of interest	400 nm (modified to 380 nm)	400 nm (modify to 380 nm)	380 nm	450-475 nm
Transparent	320-400 nm	320-400 nm	320-400 nm	320-450 (low pass) > 450 (high pass)
Reflective	400-500 nm	400-500 nm	400-500 nm	> 450 (low pass) 320-450 (high pass)
Max tranmission efficiency	> 90 % (dependent on substrate)	> 90 %	> 90 %	> 90 %
Max reflection efficiency	> 95 %	> 95 %	> 95 %	>95 %
width of edge region	<10 nm	< 10 nm	< 10 nm	< 10 nm
angle of incidence optimize	20 deg - 70 deg	20 deg to 70 deg	20 deg to 70 deg	TBD
Optimize for	45 deg	45 deg	45 deg	TBD
movement of edge within angle	< 10 nm	< 10 nm	< 10 nm	< 10 nm
Preferred Substrate	BK270 glass (Fused silica possible ?)	BK270	Still open selection	Open
	~~Try fused silica, BK270 glass, UVT acrylic, B33 glass~~	~~Try fused silica, BK270 glass, UVT acrylic, B33~~	~~Try fused silica, BK270 glass, UVT acrylic, B33~~	~~Try fused silica, BK270 glass, UVT acrylic, B33~~
Shifter	PTP on the uncoated side	PTP on uncoated side	PTP on uncoated side	No shifter is needed on any filter.

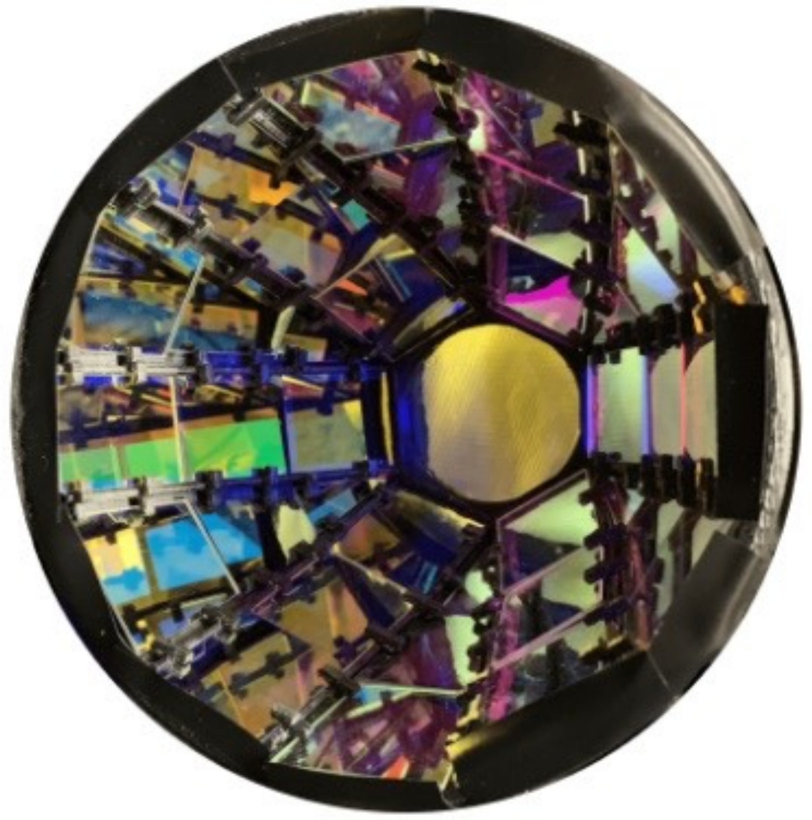
DUNE

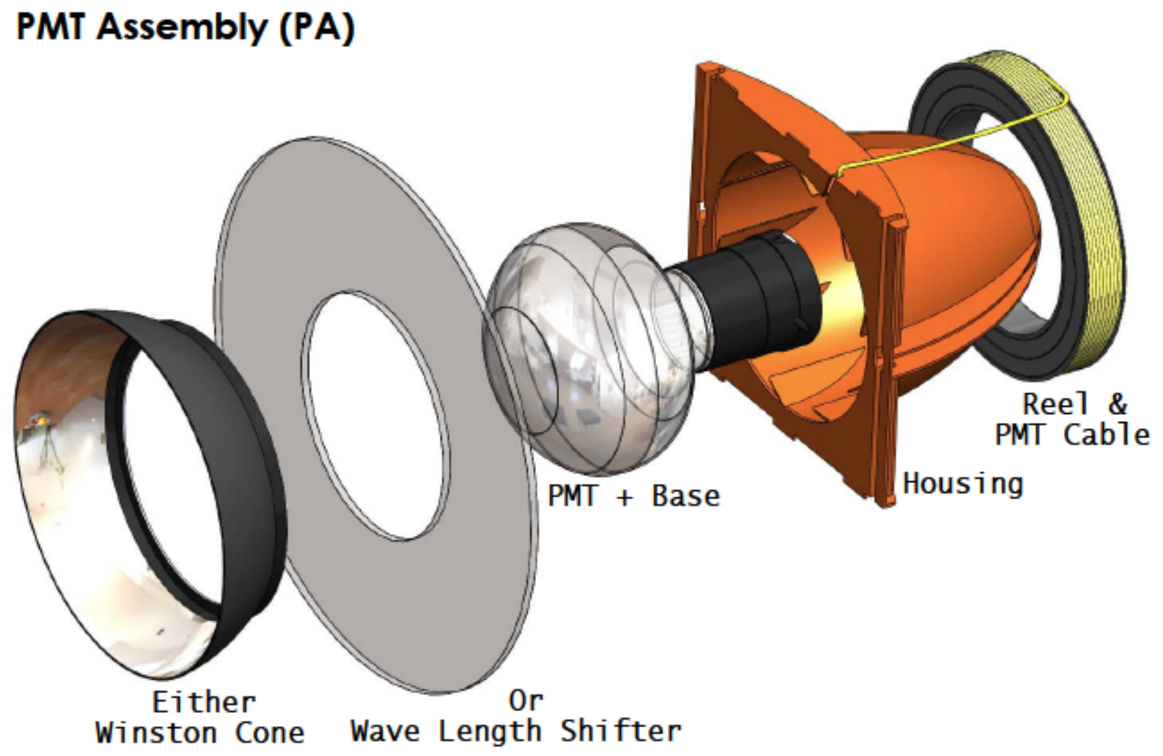
Liquid Argon ARAPUCA detector concept and roles of DFs

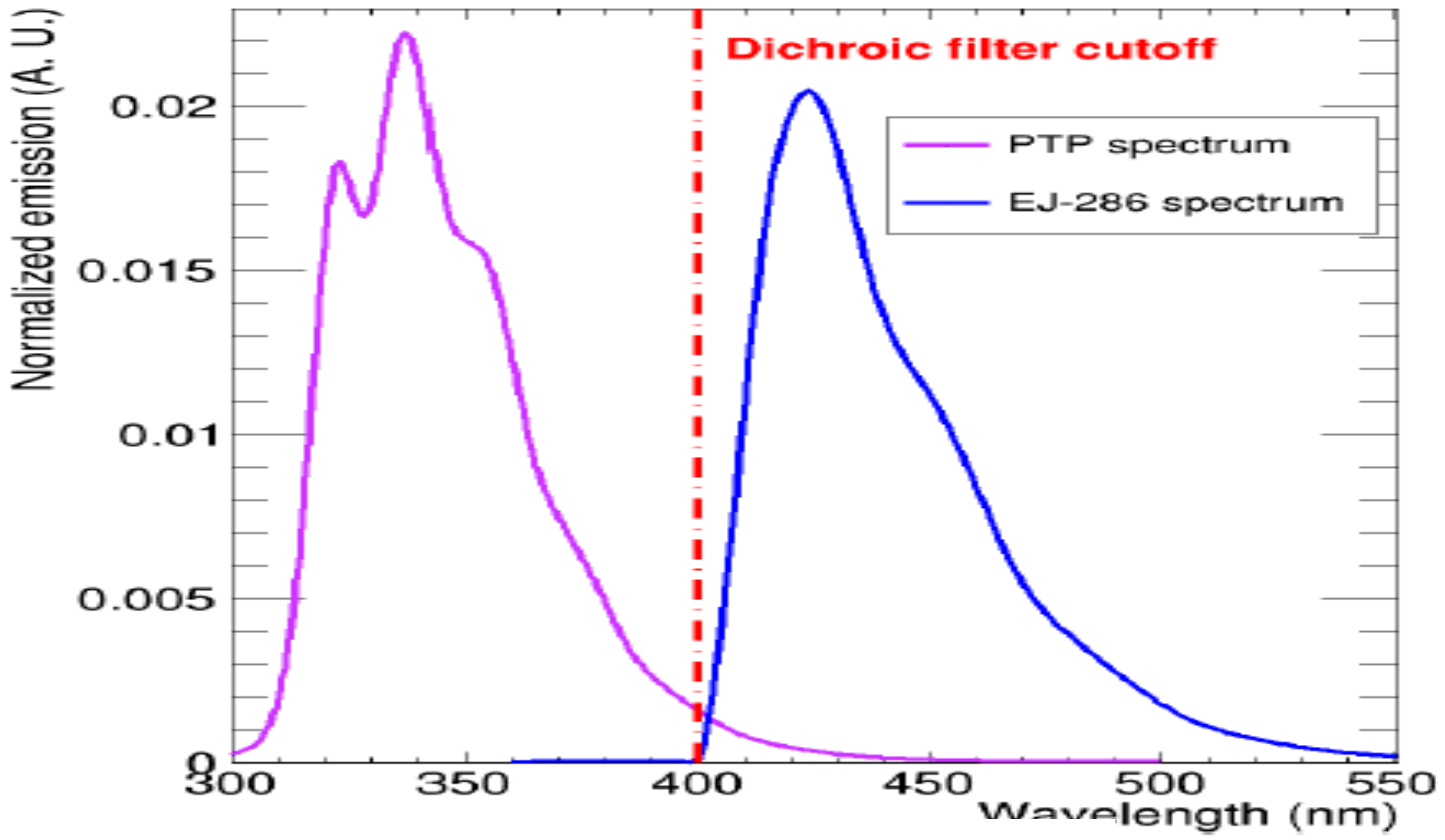
THEIA

Winston cone(WC) /wavelength shifting plates(WLSP) type of detector concept and roles of DFS

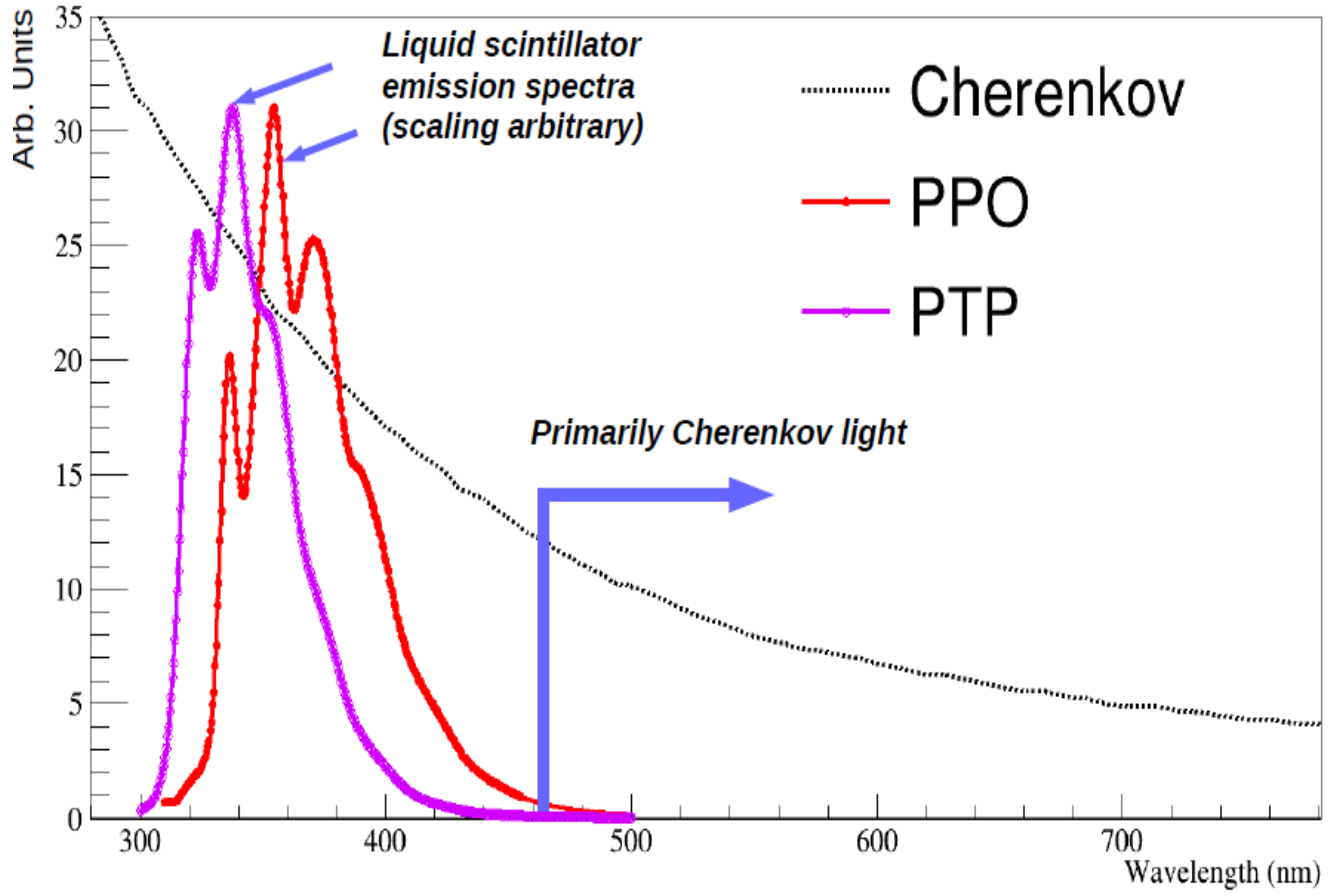




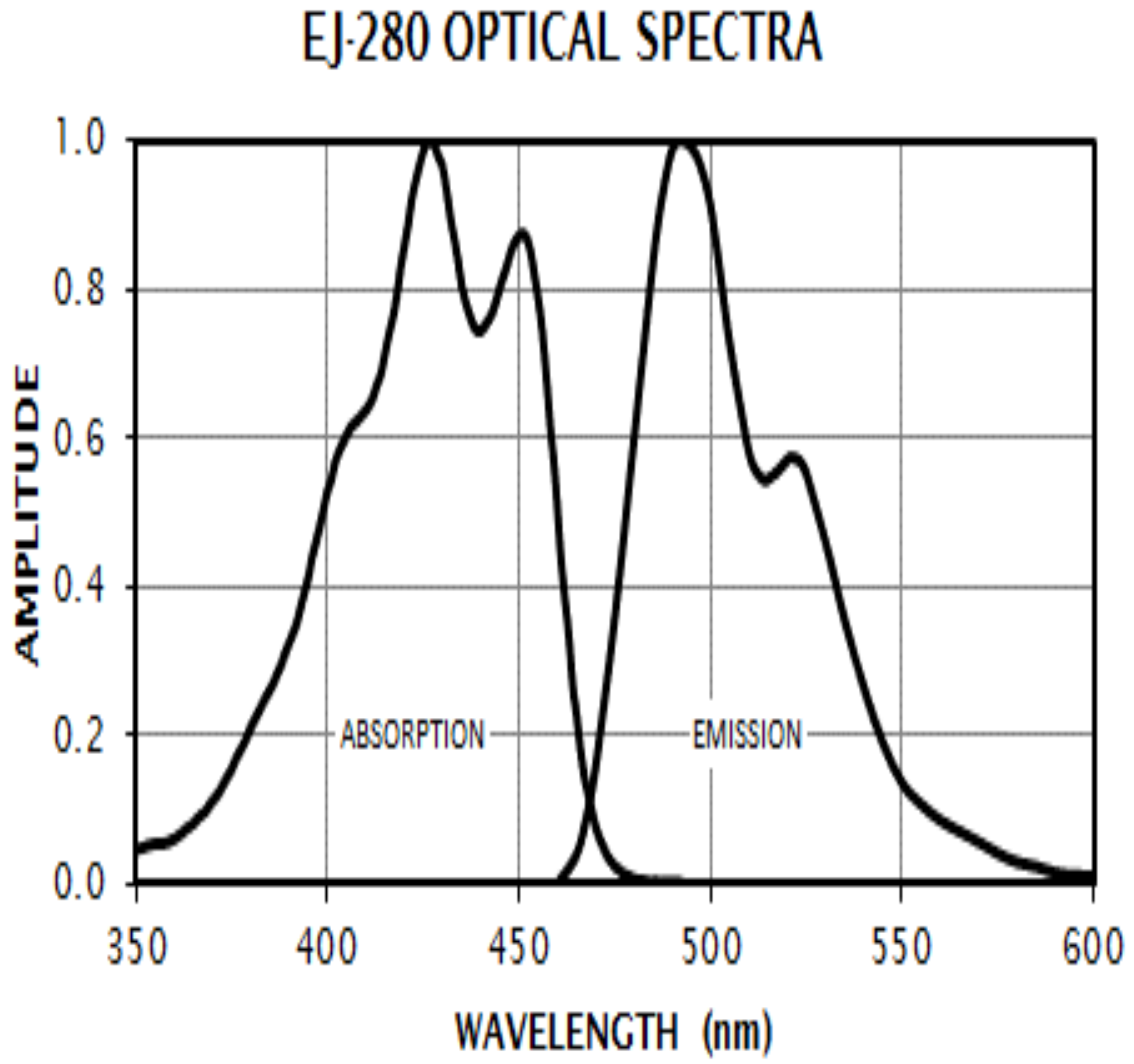




(Polymer EJ-286 WLSP)



Goal is to achieve Cherenkov and scintillation separation while losing as few total photons as possible.



The dichroic cutoff (red dashed line), the PTP (purple) and the EJ-286 emission spectra. (b) X-ARAPUCA principle of work, with total internal reflection and the reflective cavity trapping photons.

The SP filters tile the barrel of the Winston cone and a central LP filter is placed at the aperture. A small amount of black electrical tape is used to block a small gap between the filters and the holder at the top of the dichroicon.

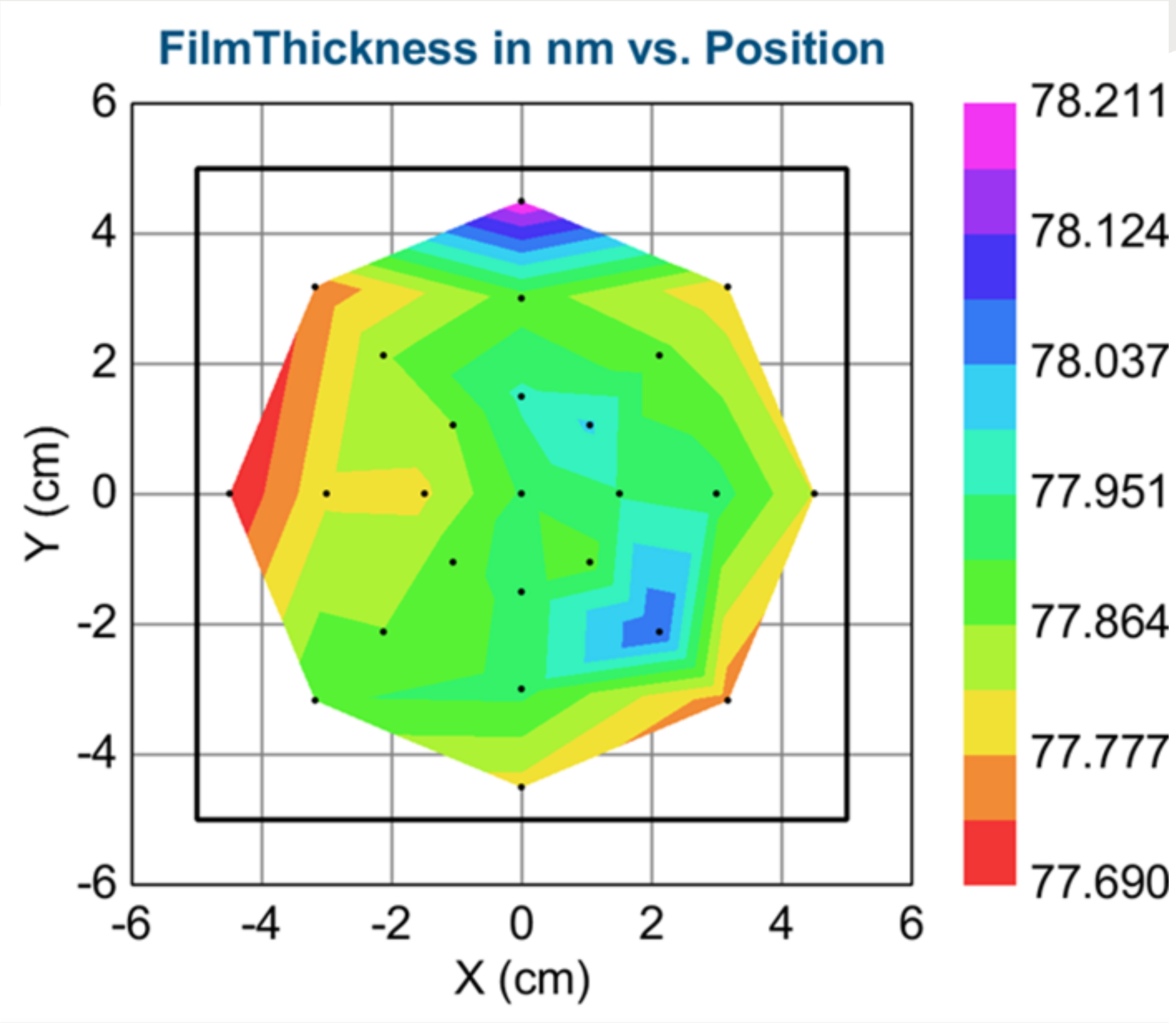
Significant Technology Breakthrough from Raytum Photonics, What has been achieved.

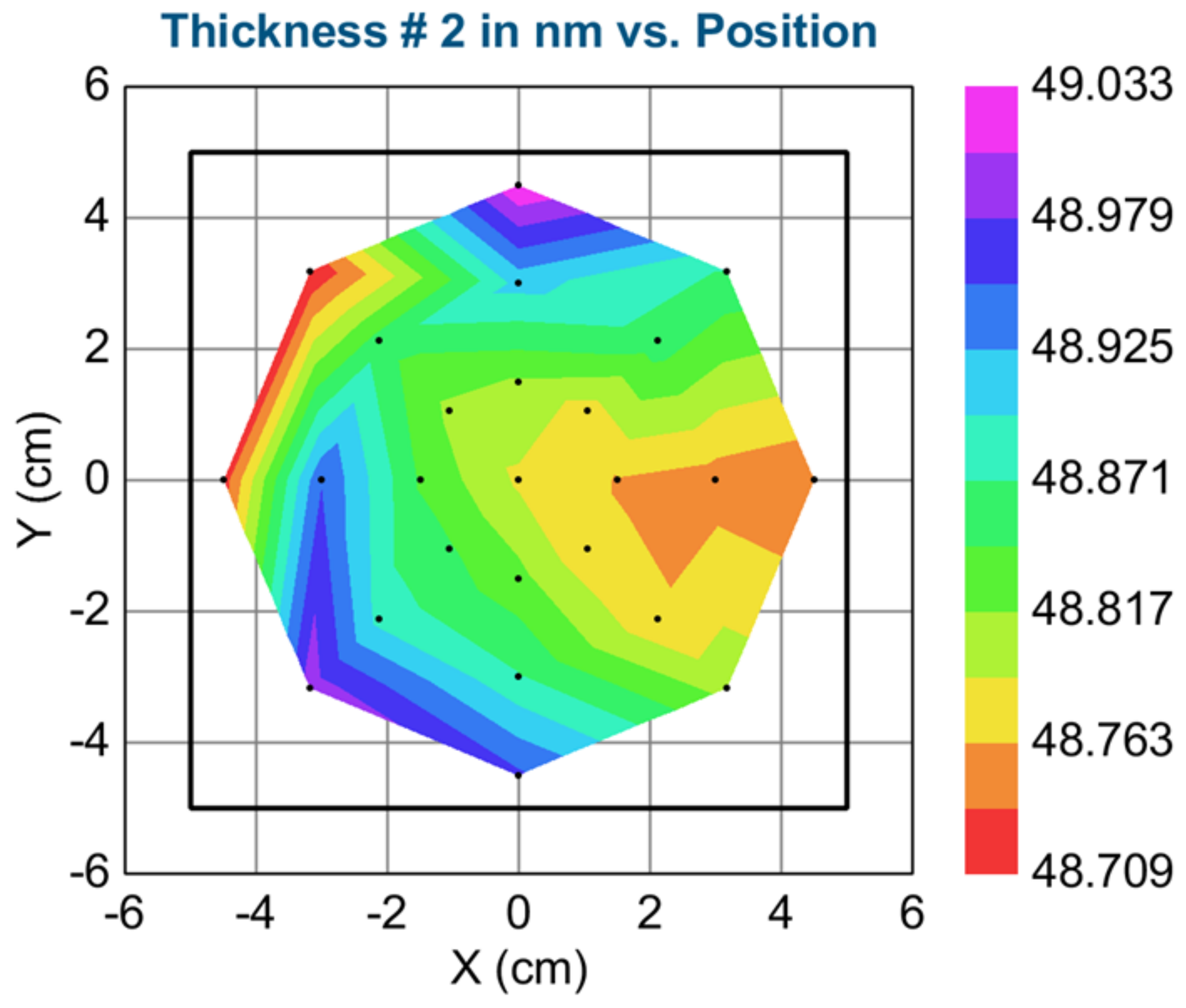
RAYTUM Photonics

- Large Area Coating using ALD Technology has been achieved. Long pass filters in UV band are successfully demonstrated over different substrates. The results match with the design very well.
- Extremely low absorption, only tens of ppm for 68 layers of coating , was confirmed by PCI technique.
- The uniformity as low as 0.12% has been achieved.
- By optimizing the coating process, the production rate could be greatly improved.
- The fabrication of short pass filters been performed. And I will provide some results.

Coating Uniformity Measurement

RAYTUM Photonics





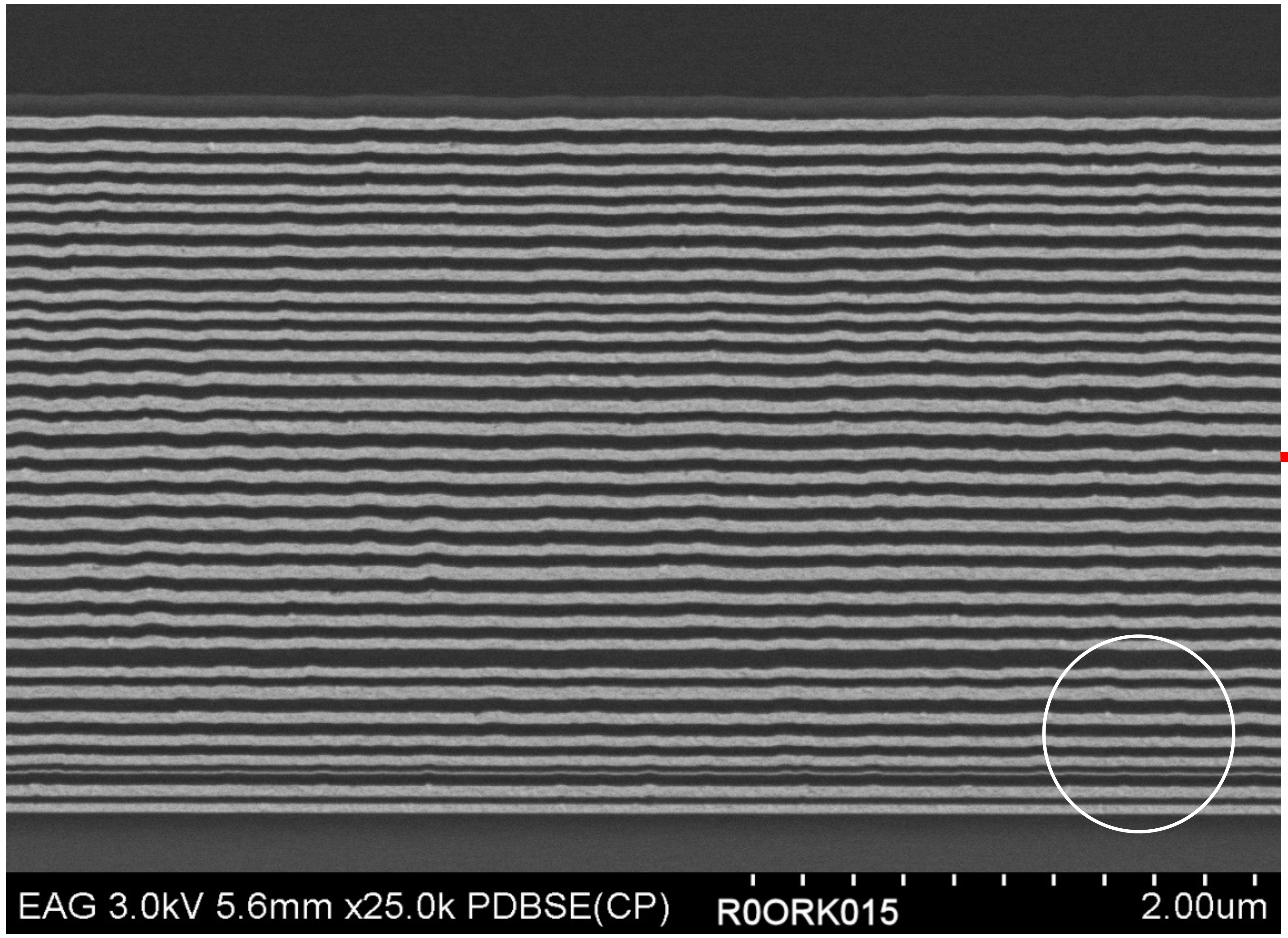
Parameter	Average	Std. Dev.	Slope	Min	Max	Range
Thickness in nm	77.88561	0.10829	0.12%	77.69048	78.21082	0.52035

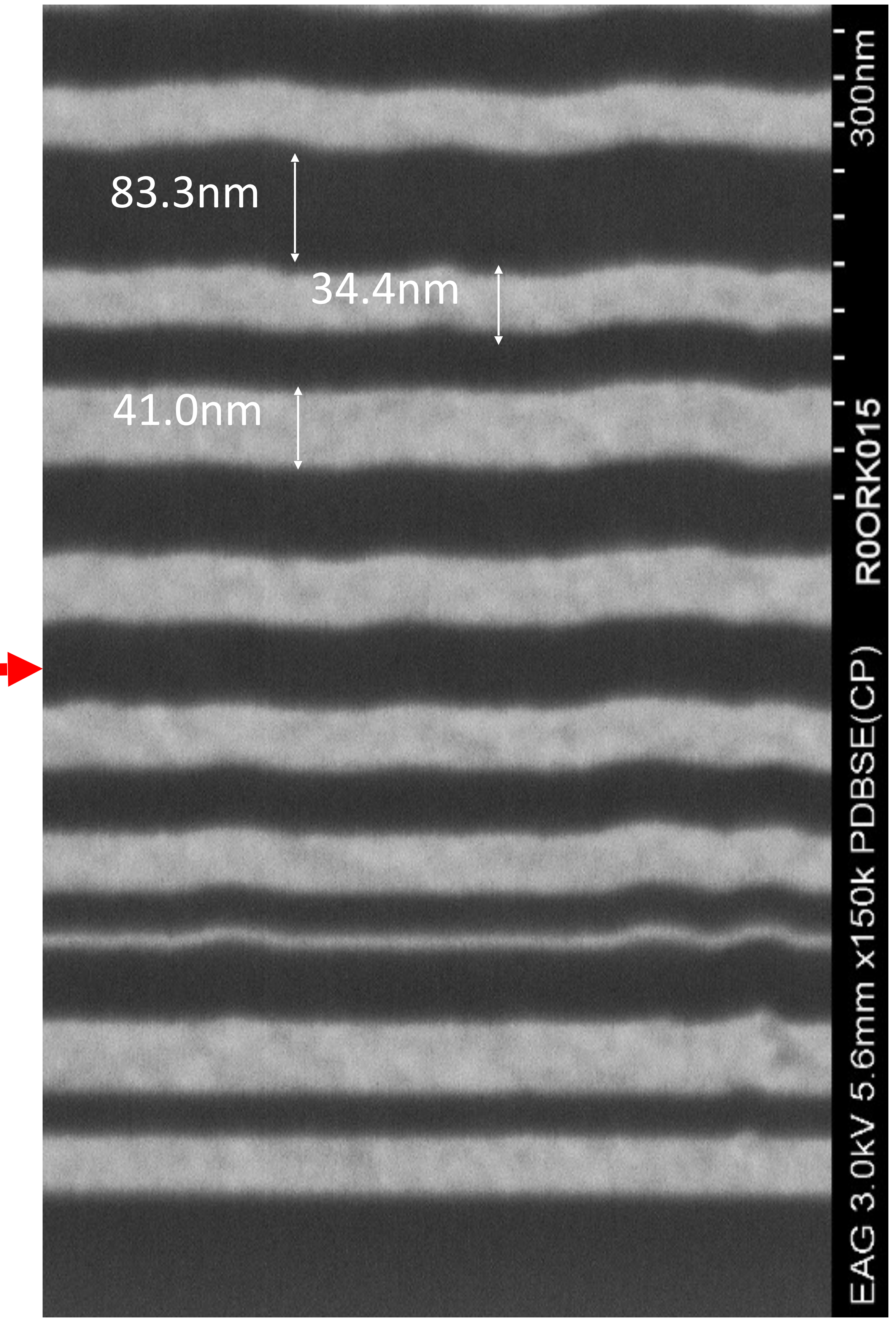
Parameter	Average	Std. Dev.	Slope	Min	Max	Range
Thickness in nm	48.83677	0.08485	0.17%	48.70925	49.03308	0.32383

Low Index Material

High Index Material

Cross sectional SEM imaging of a full long pass Dichroic filter consisting of 64 total layers





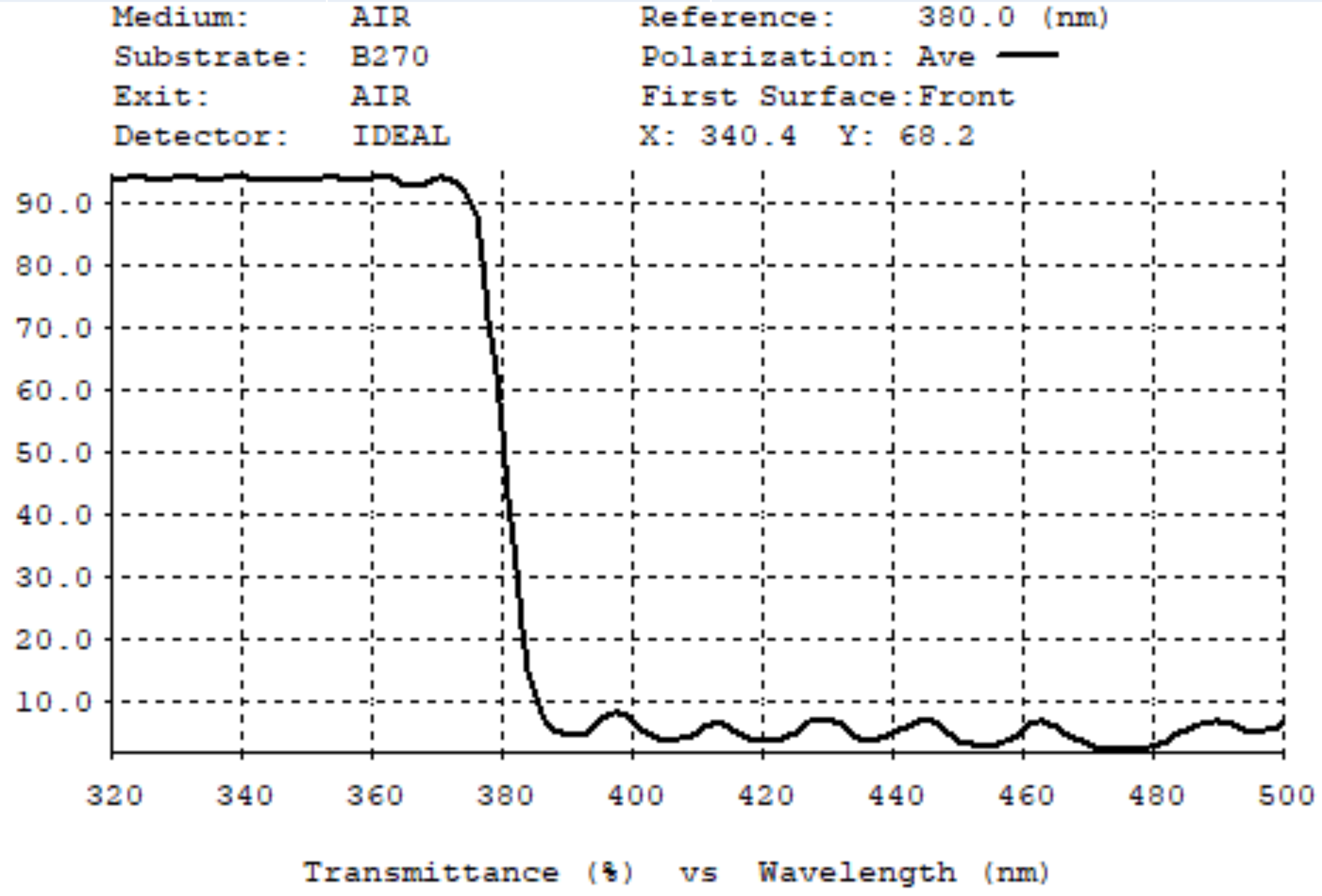
Dark band: dielectric material #1

Light band: dielectric material #2

Measurements are calibrated by a standard sample, estimated measurement error (±1nm).

New SP filters coated by S200 at Raytum

Serial Number	Substrate	Run	ALD method	deposition temperature(°C)	Material	Comment
A1	Sapphire, B33, B270(100mmx77mm)	4	Thermal temporal	200	highly tensile HfO2 by nanolaminate /Al2O3 tensile	no cracks o sapphire, visible crack lines on B270, B33

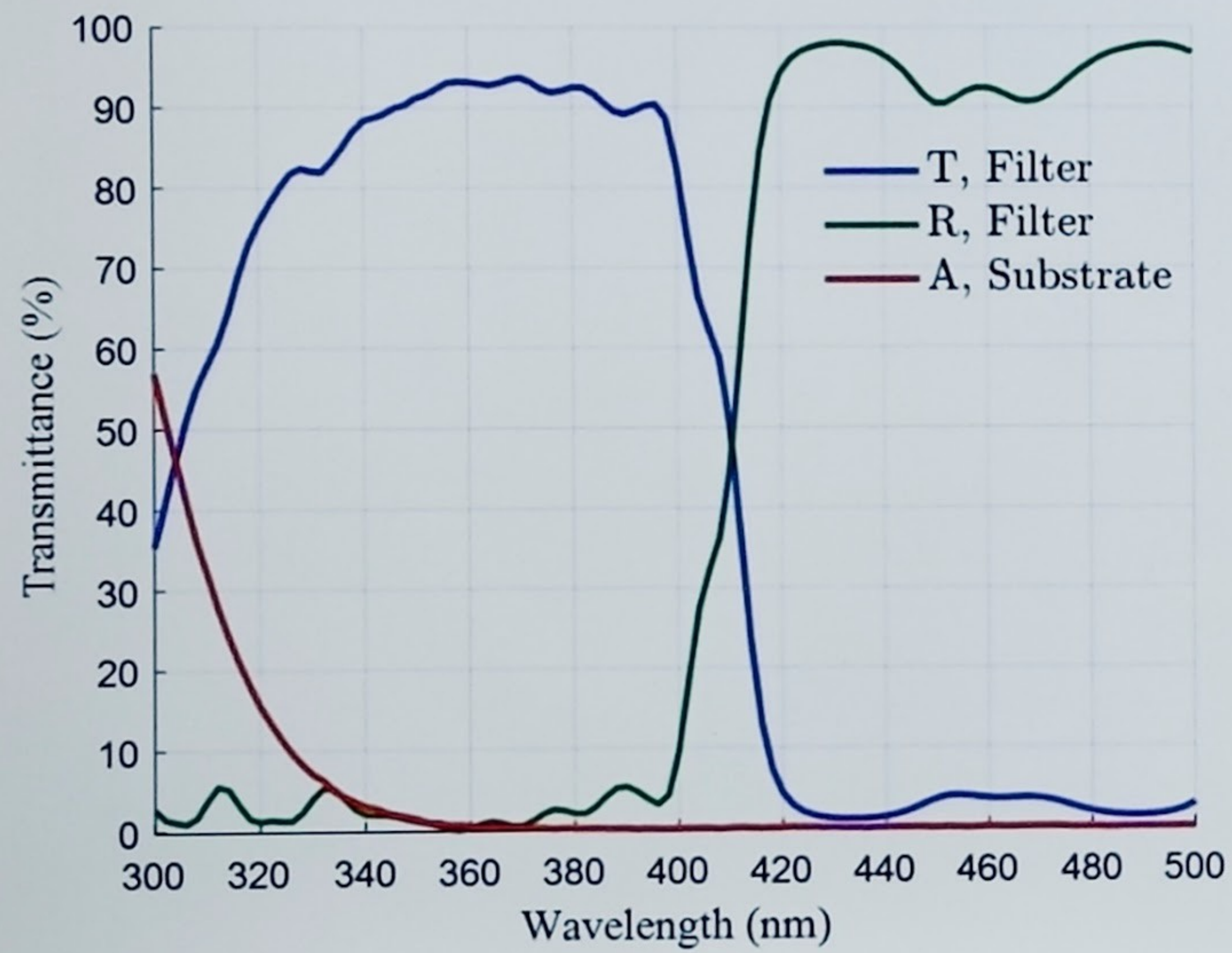


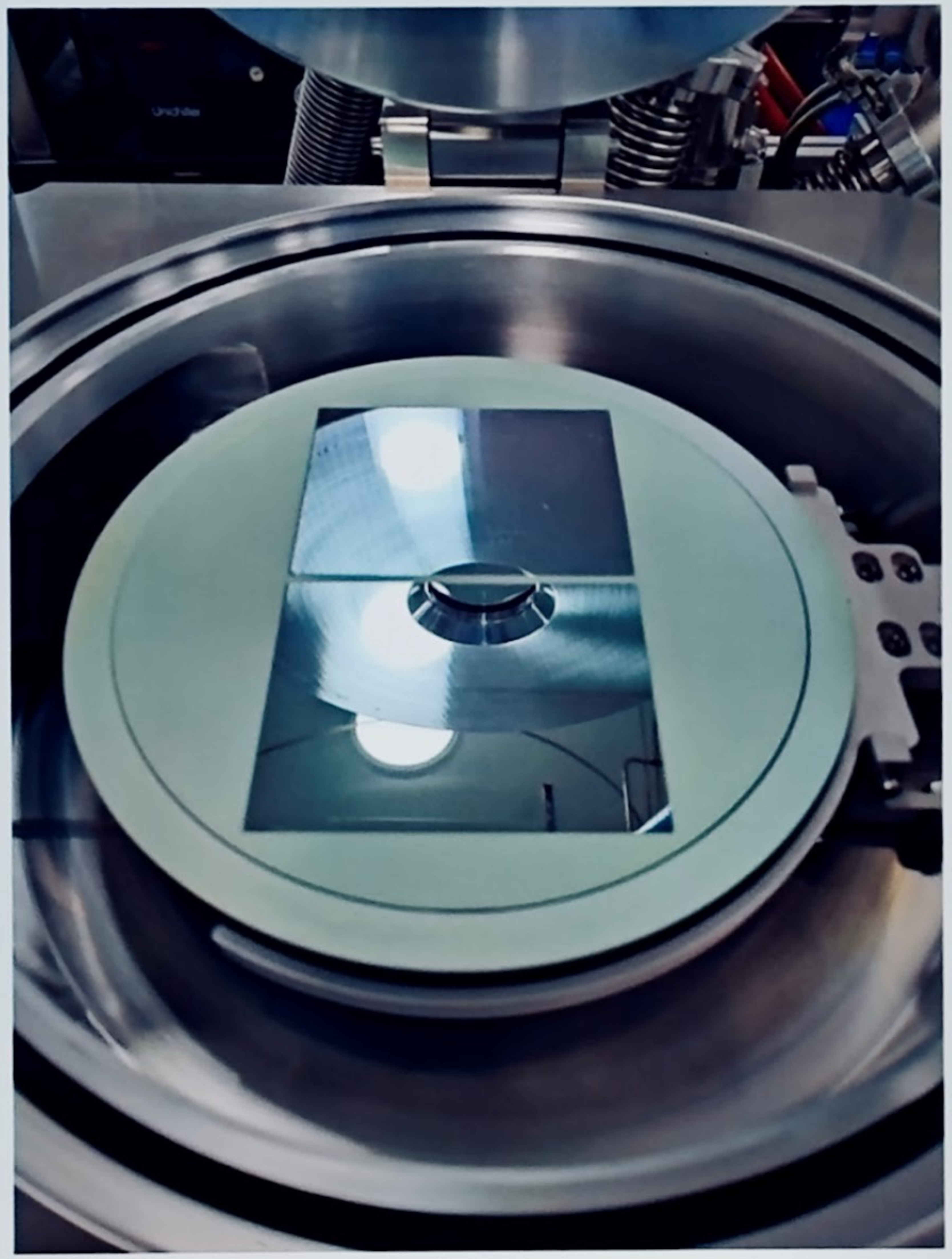
Cut wavelength 380 nm

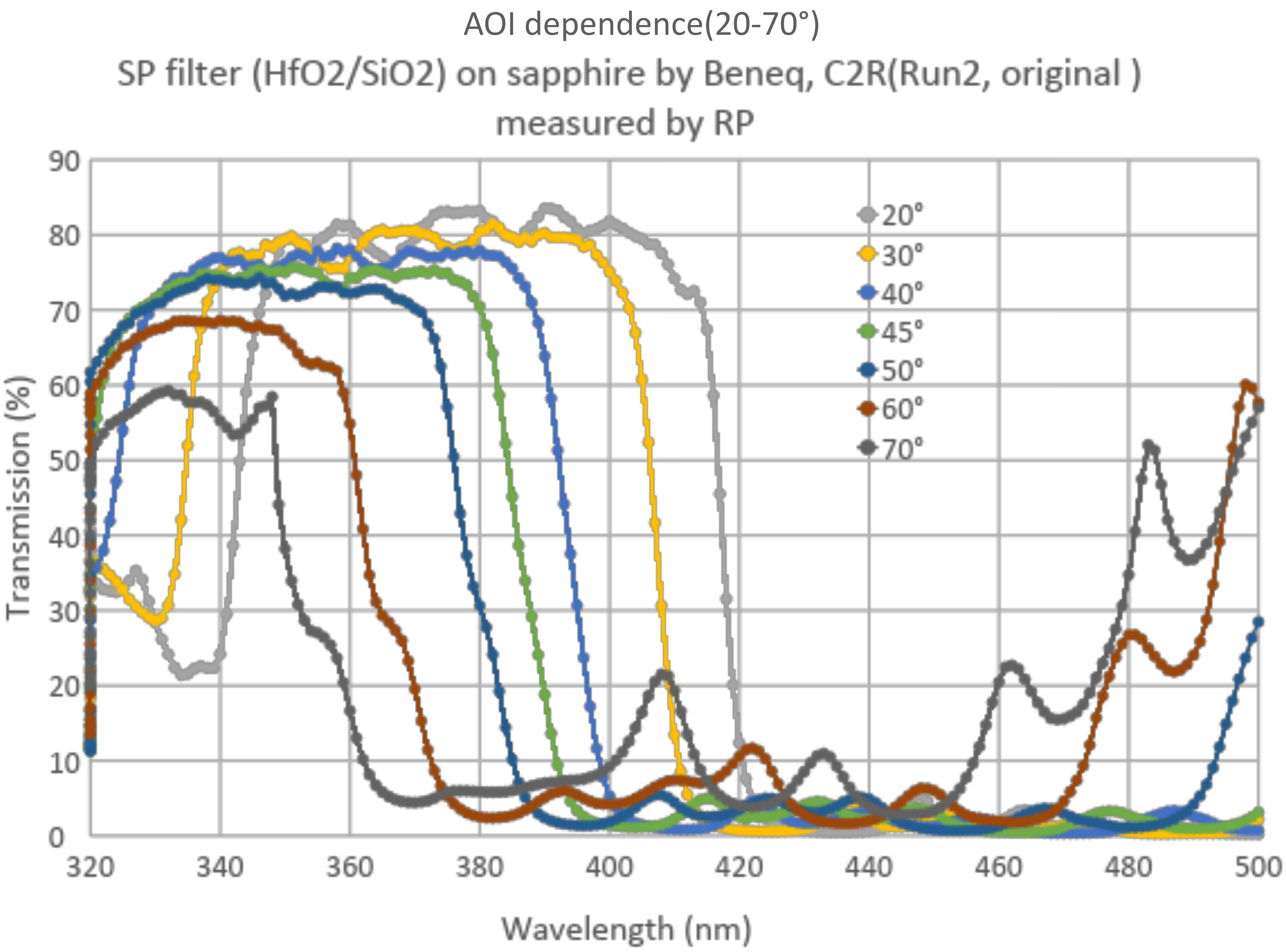
1. Use nanolaminate structure(NL) to prevent formation of crystals at 200°C and reduce Al2O3 internal compressive stress.
2. Cutting-off edge tuned to 380nm in order to prevent leak of back scattered WLS light at 430nm(peak) at AOI from 20-70°.
3. Use different substrates to verify UV transparency.(sapphire, B33, B270)
4. Optimized in air not in liquid AR or water.
5. AOI dependence(20-70°) is studied on cutting edge width(spec<10nm), and it seems that very limited improvement thus decided to go ahead with#1-4.

Preliminary Results

- SiO_2 and HfO_2 at 100°C
  - Both films amorphous, stress-controlled processes
  - Preliminary results on B270-substrates
  - Design is 37-layers, 2900 nm thick

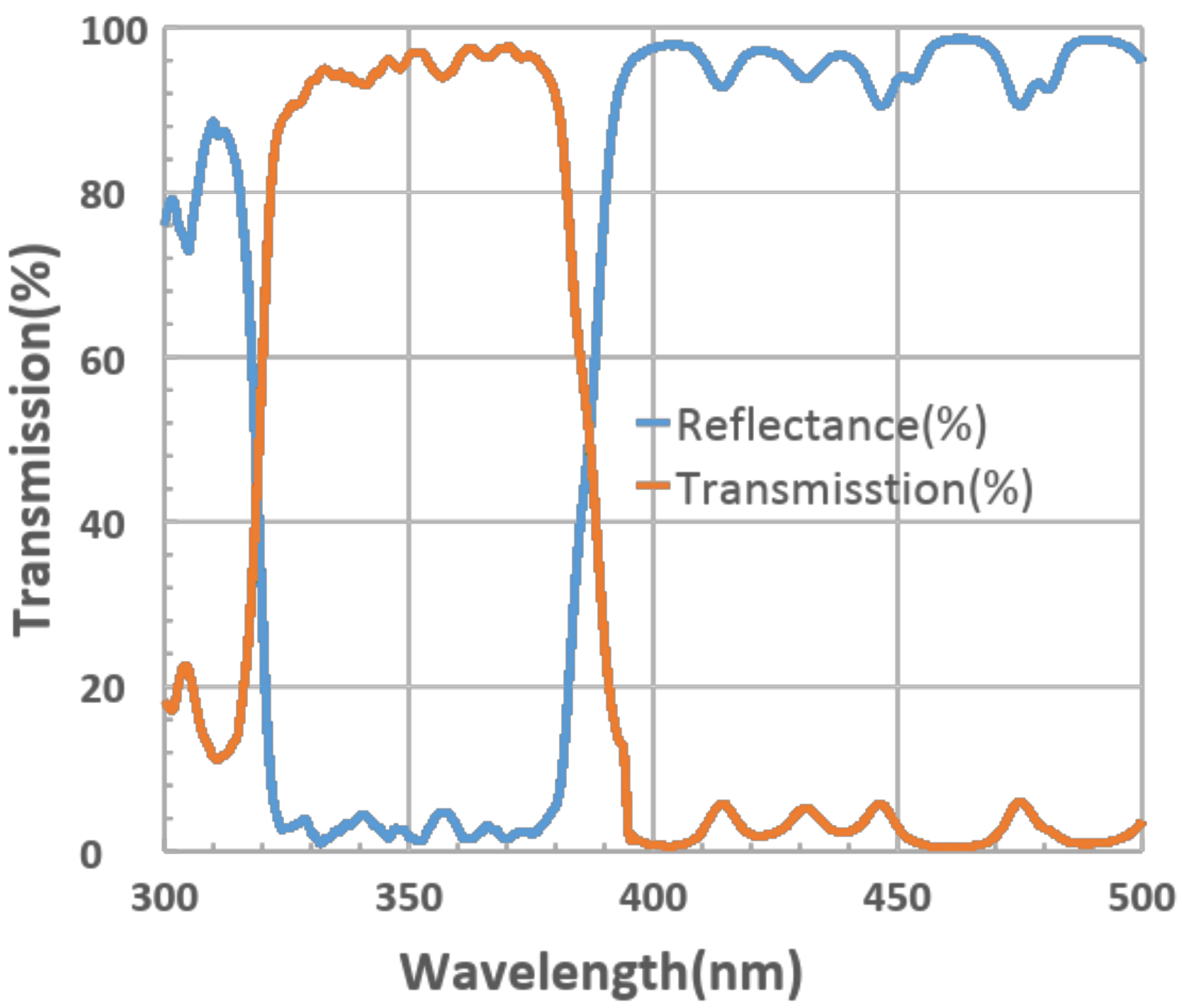






1. Edge width(15nm)
2. Edge movement at AOI 20-70°(70-80nm)

Run2



This is the most recent fabrication run of a SP filter on 100mmx77mm sapphire by the Beneq C2R coater.

1.Transmission is recalibrated for sapphire both side reflectance.(confirmed by PR)

2.Transmission in UV < 340nm is significantly improved by using a sapphire substrate. This proves that the it is the substrate not the SP filter itself, that limits transmission near UV region.

3.The cutting-off edge is tuned from 400nm to 380nm which can significantly reduce reflected light >410-420nm from the WLSP (EJ286) and improve the angular dependence.

BENEQ C2R coater

ALD is now extremely important technology for silicon production.

The new gate structures for near future GPU's will be manufactured using ALD for part of the process.

The key leader in this is https://beneq.com/en/

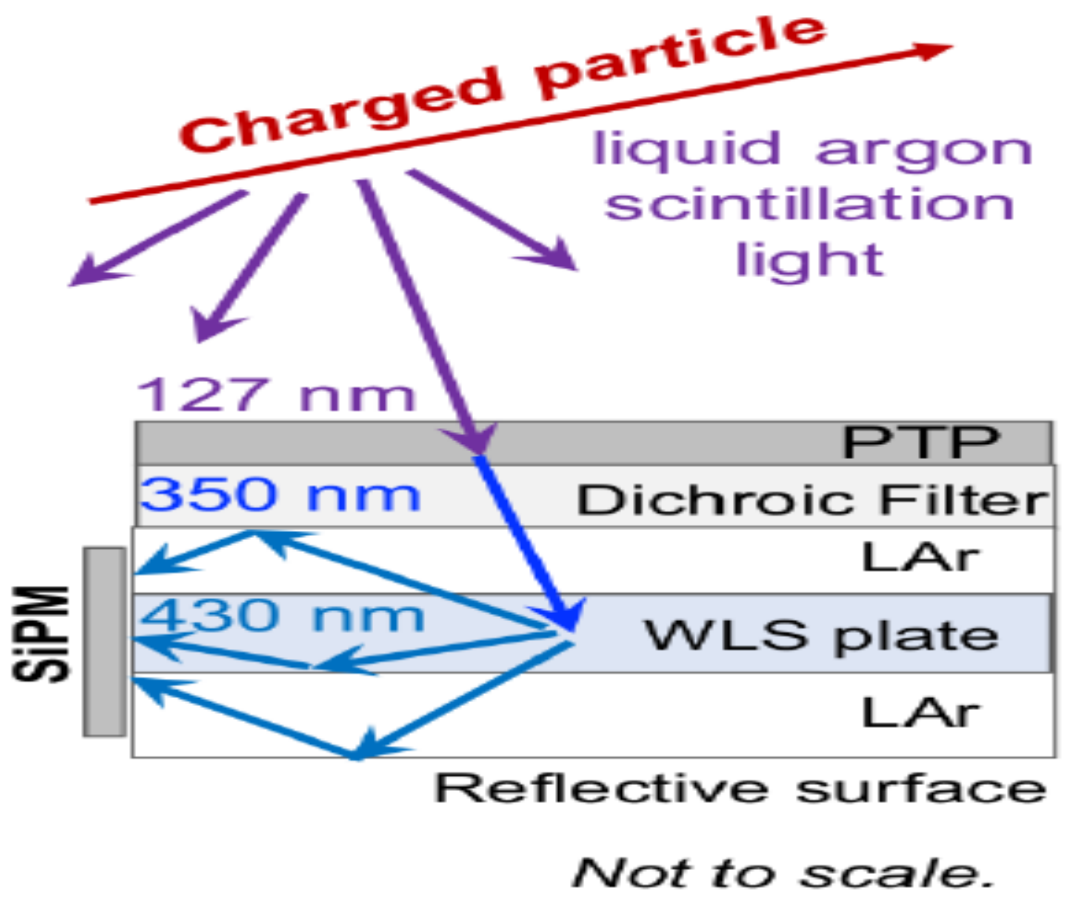
The filters delivered from RAYTUM were made with BENEQ system C2R

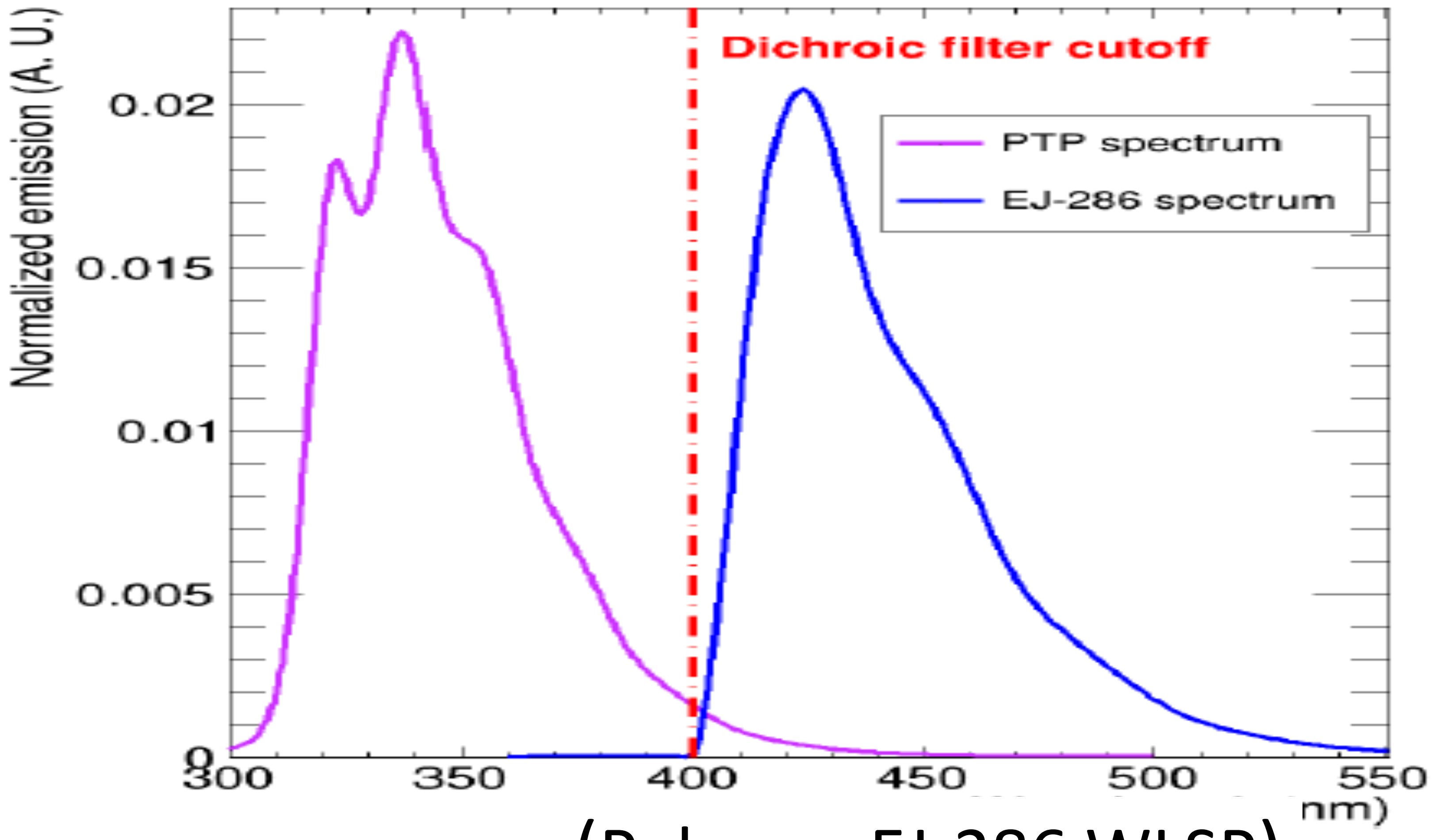
"Beneq C2R provides an optimal solution for high performance ALD in industrial applications, such as optical coatings and barriers."

Refined design model considering light back scattering from WLS plate
(considering both AOI angle distribution of 20-70° and wavelength distribution)

DUNE

Liquid Argon ARAPUCA detector concept and roles of DFs

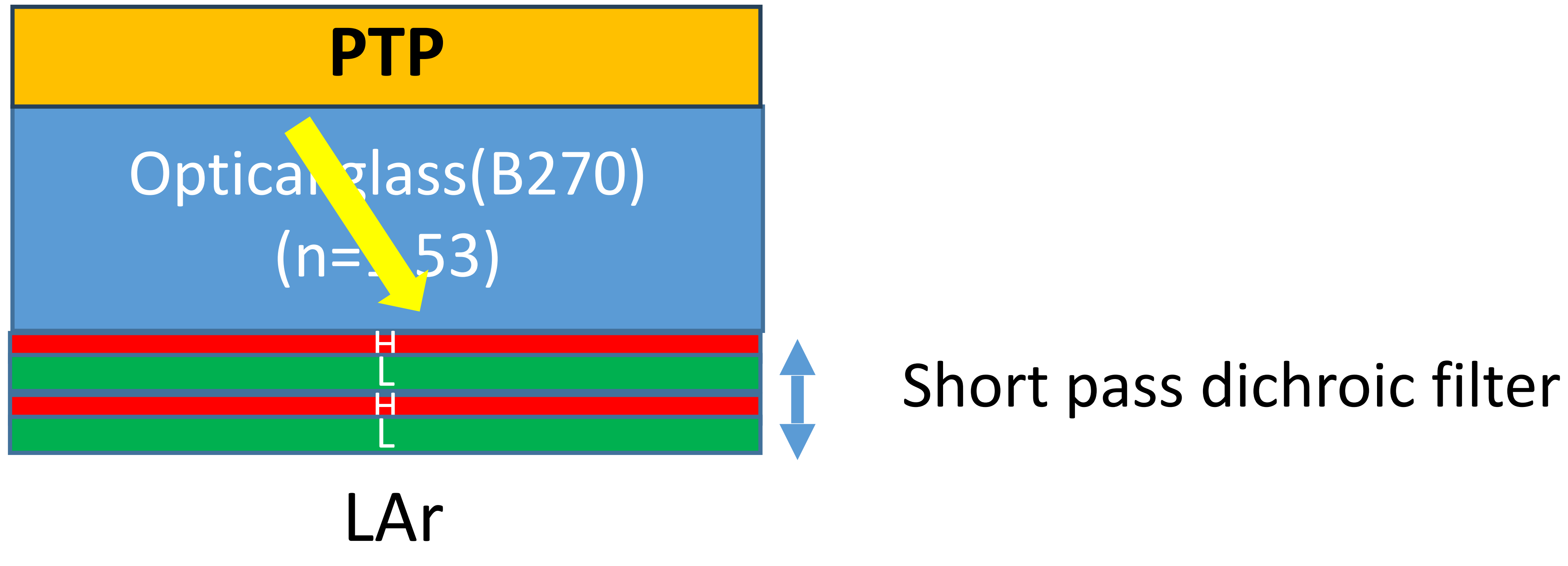


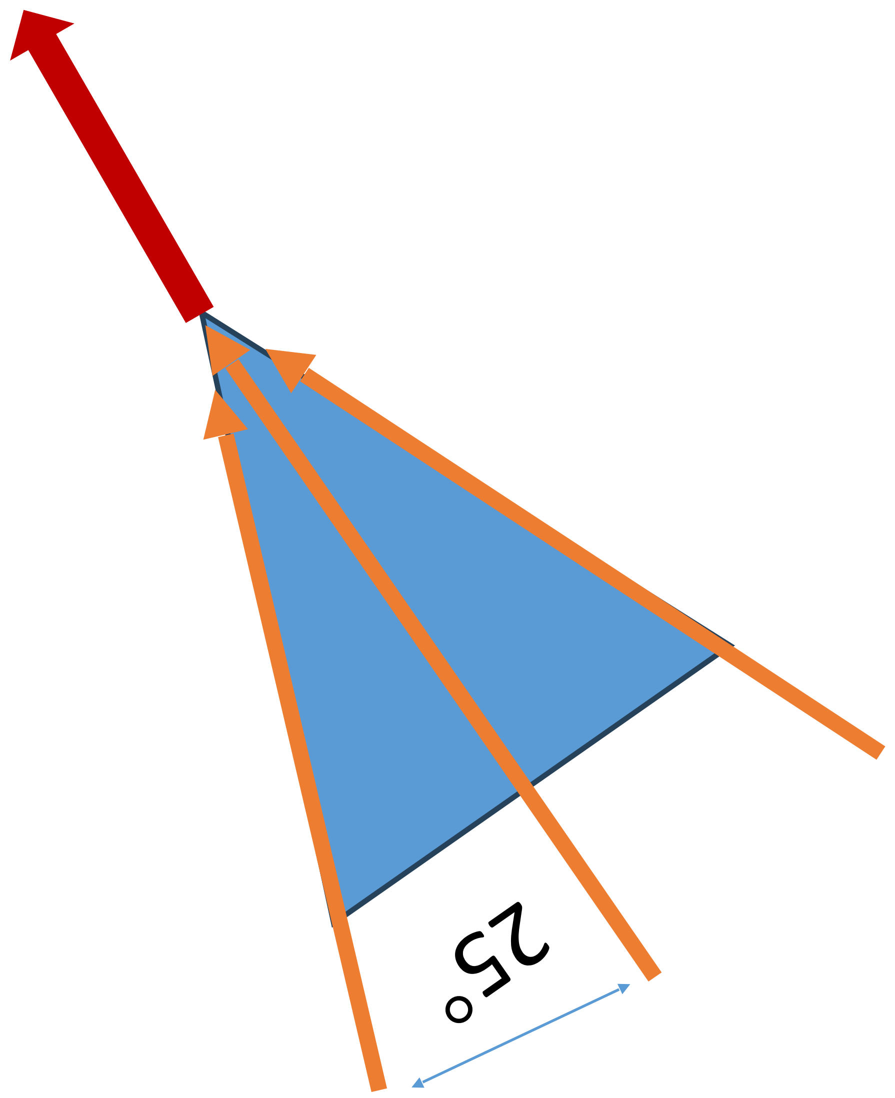


(Polymer EJ-286 WLSP)

The dichroic cutoff (red dashed line), the PTP (purple) and the EJ-286 emission spectra. (b) X-ARAPUCA principle of work, with total internal reflection and the reflective cavity trapping photons.

Incident angle of light(AOI)=45° with no AOI distribution(λ=350 nm)





AOI with 20-70° distribution profile undefined yet(λ=430 nm)(Equal or Cosine Or Gauss)

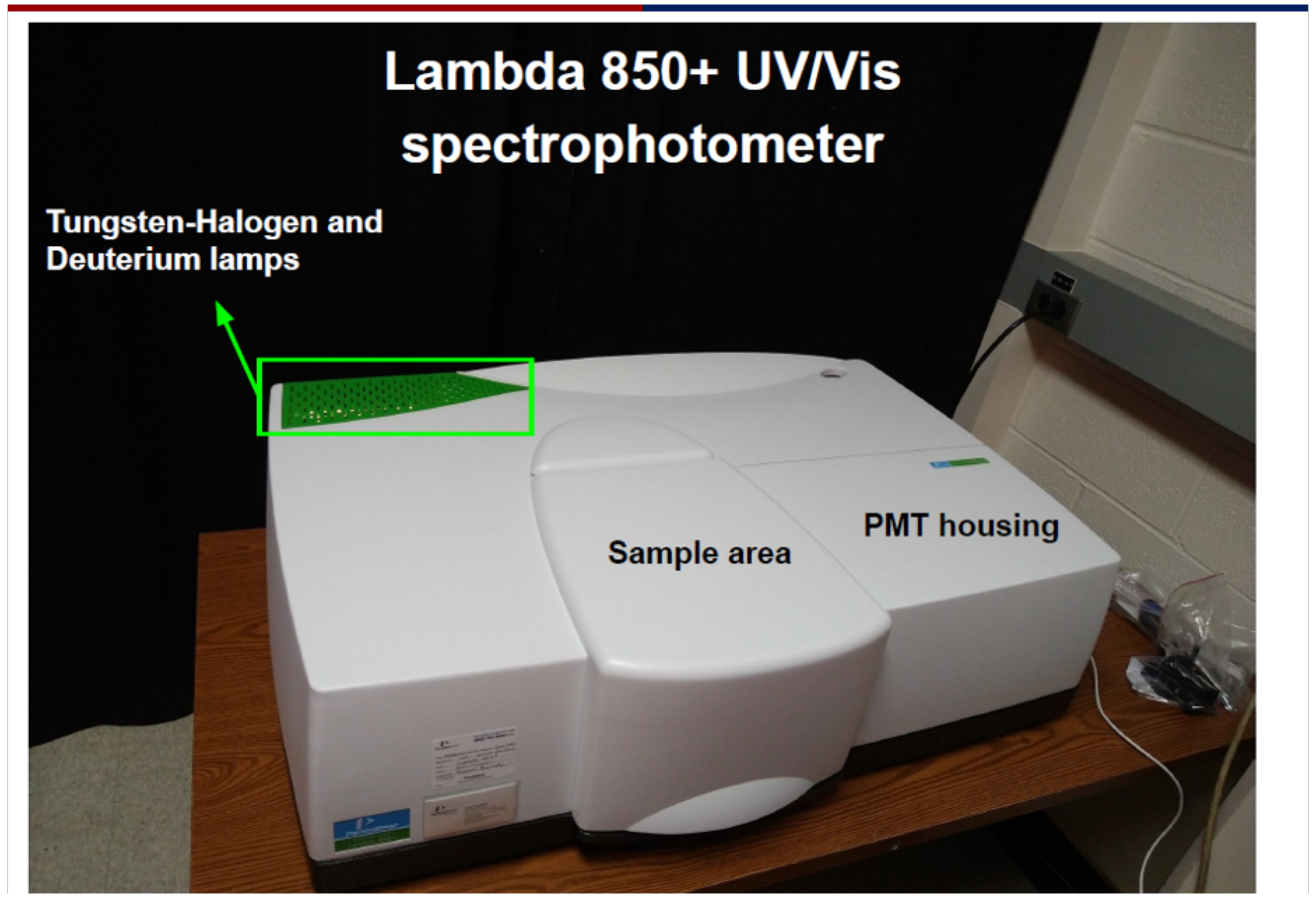
Short Pass Filters Delivered and measurements

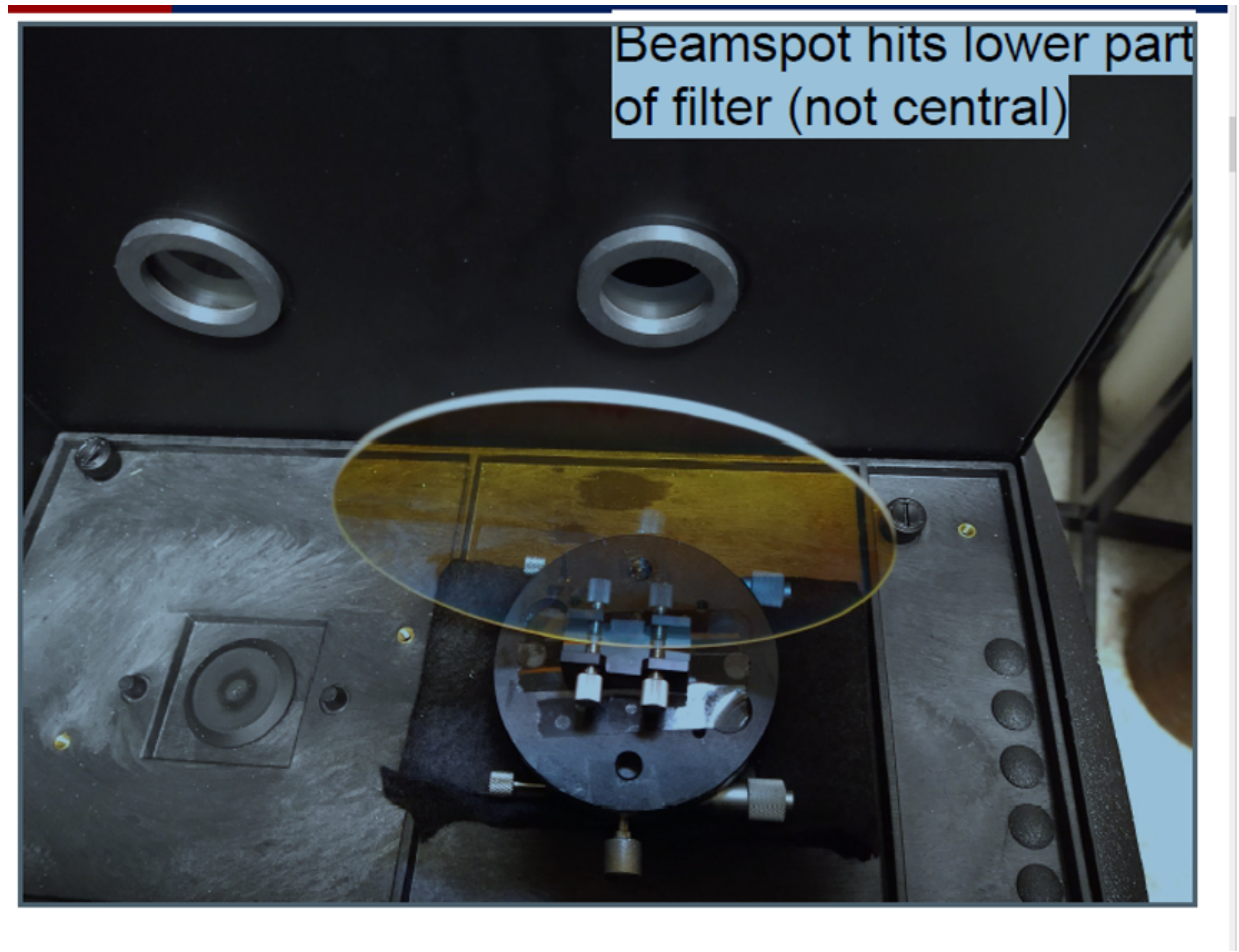
4 delivered to BNL and 3 to UPenn.

Number	Substrate		Size	Cut wavelength	location
B16	Sapphire	HfO_2/Al_2O_3	77 x 100 x 1 mm	380 @ 45 deg	BNL
B17	Sapphire	HfO_2/Al_2O_3	77 x 100 x 1 mm	380 @ 45 deg	BNL
B20	B33	HfO_2/Al_2O_3	77 x 100 x 1 mm	380 @ 45 deg	BNL
B21	B33	HfO_2/Al_2O_3	77 x 100 x 1 mm	380 @ 45 deg	BNL
NA	Sapphire	HFO_2/Al_2O_3	4 inch circle x 1 mm	380 @ 45 deg	UPenn
B23	B33	HFO_2/SiO_2	77 x 100 x 1 mm	380 @ 45 deg	UPenn
B15	Sapphire	HFO_2/SiO_2	77 x 100 x 1 mm	380 @ 45 deg	UPenn

measurements

At penn and BNL

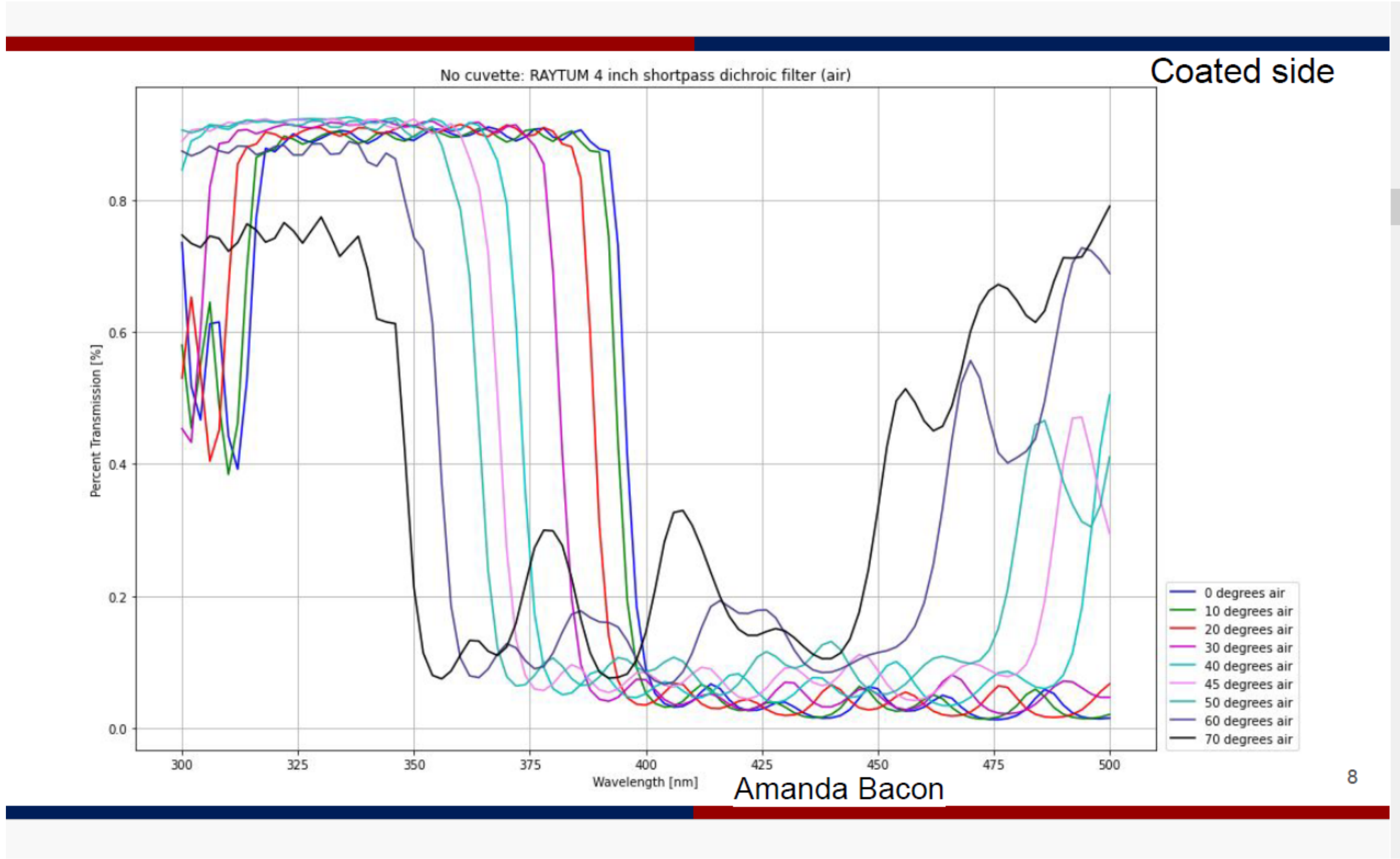


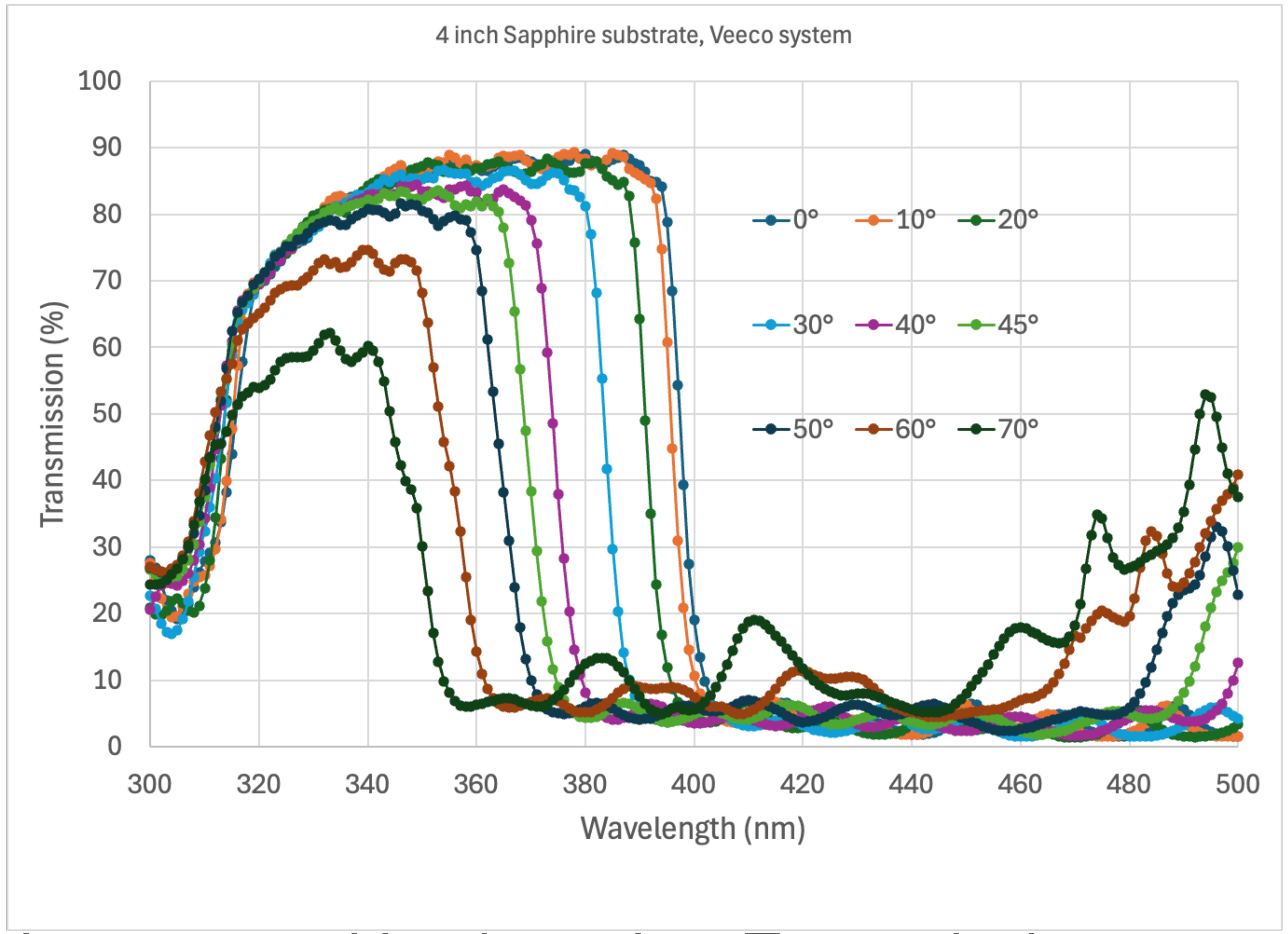


Shimadzu 1900

- Measurements were done in air.
- add 8 % for backside reflectance since this is in air.

UPENN 4 inch measurement compared to RAYTUM



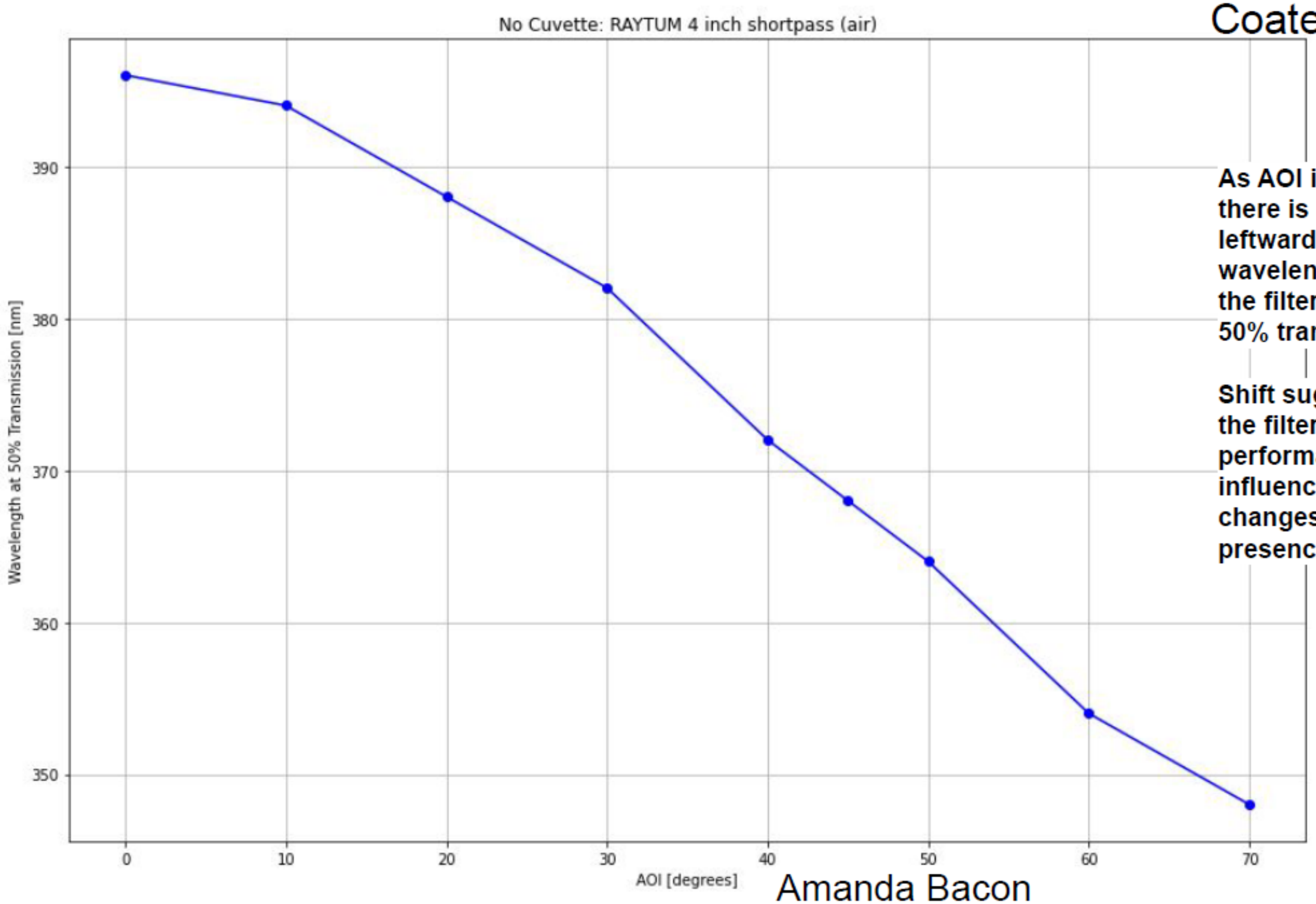


This is on Sapphire. The backside reflectance needs to be corrected in air, and so Transmission band is almost 100%

Optimized for 45 deg.

Coatings are HfO2/AL2O3

Coated side



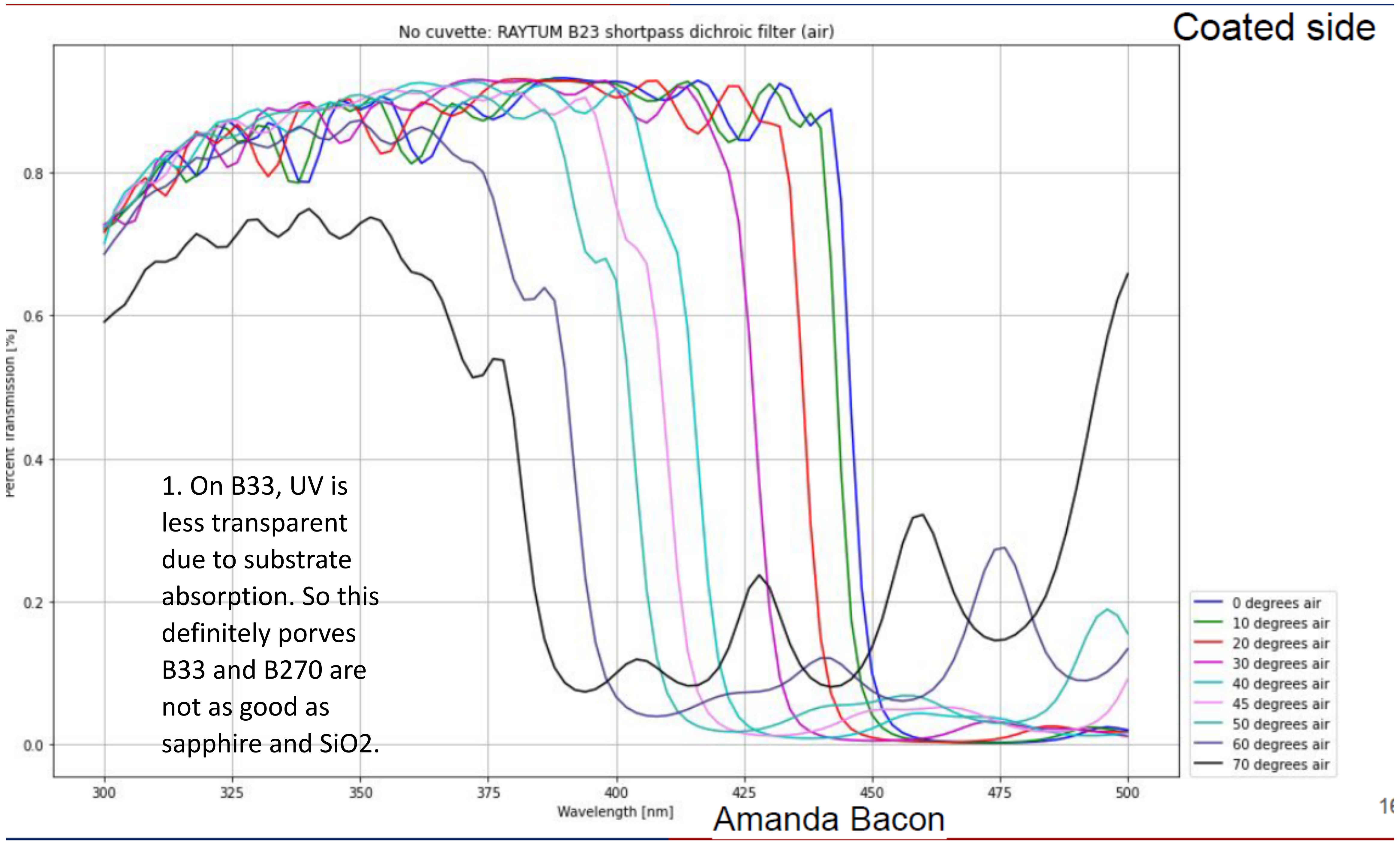
As AOI increases, there is a noticeable leftward shift in the wavelength at which the filter achieves 50% transmission.

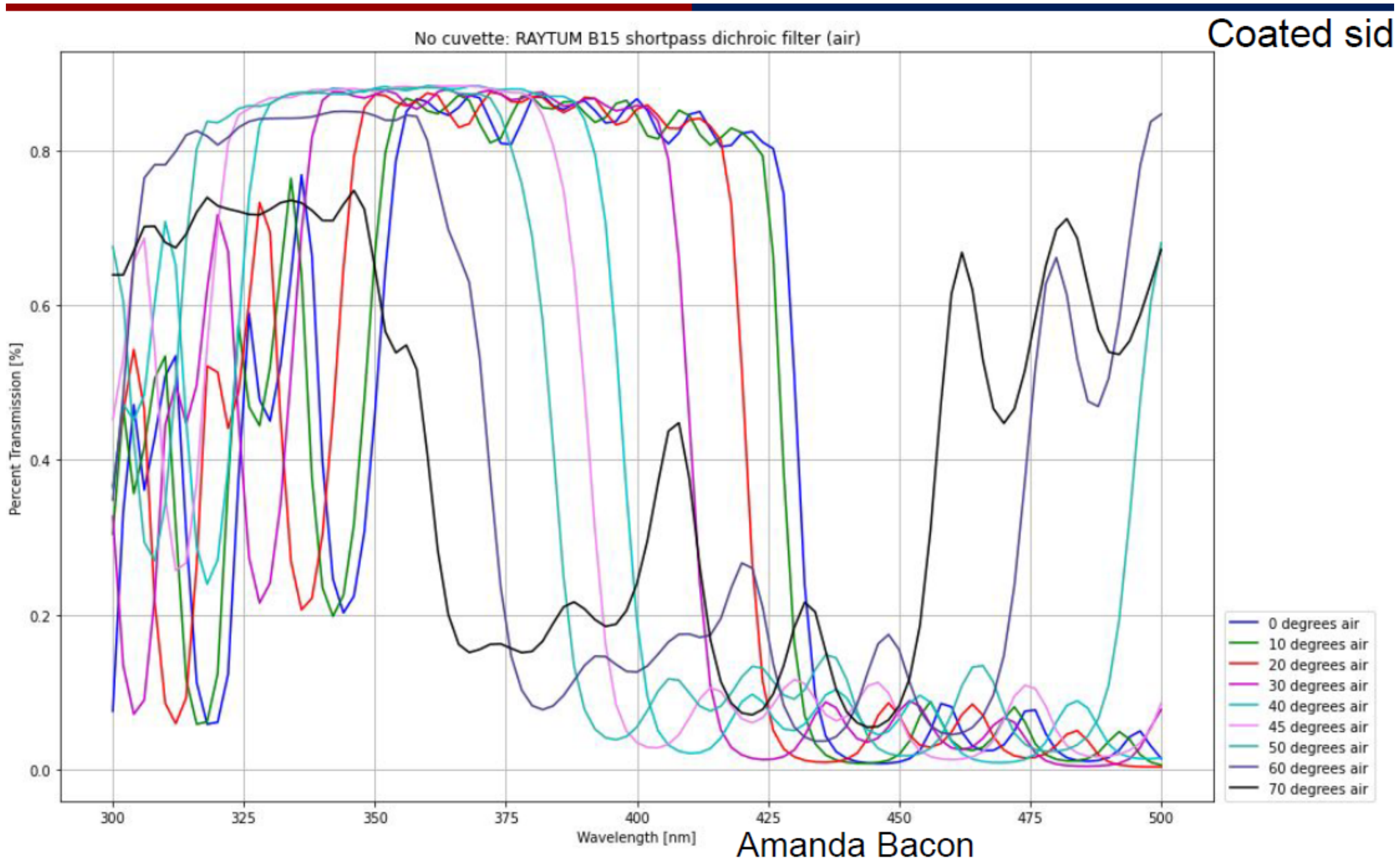
Shift suggests that the filter's performance is influenced by changes in AOI in the presence of air.

Amanda Bacon

Edge shift=50nm(AOI=0-70°) on sapphire with HfO_2/Al_2O_3 SP DF

UPENN B23 and B15 measurements on other samples





B33 glass (B23 sample) UV performance not as good due to substrate absorbance. B33 coating is HFO2/SiO2

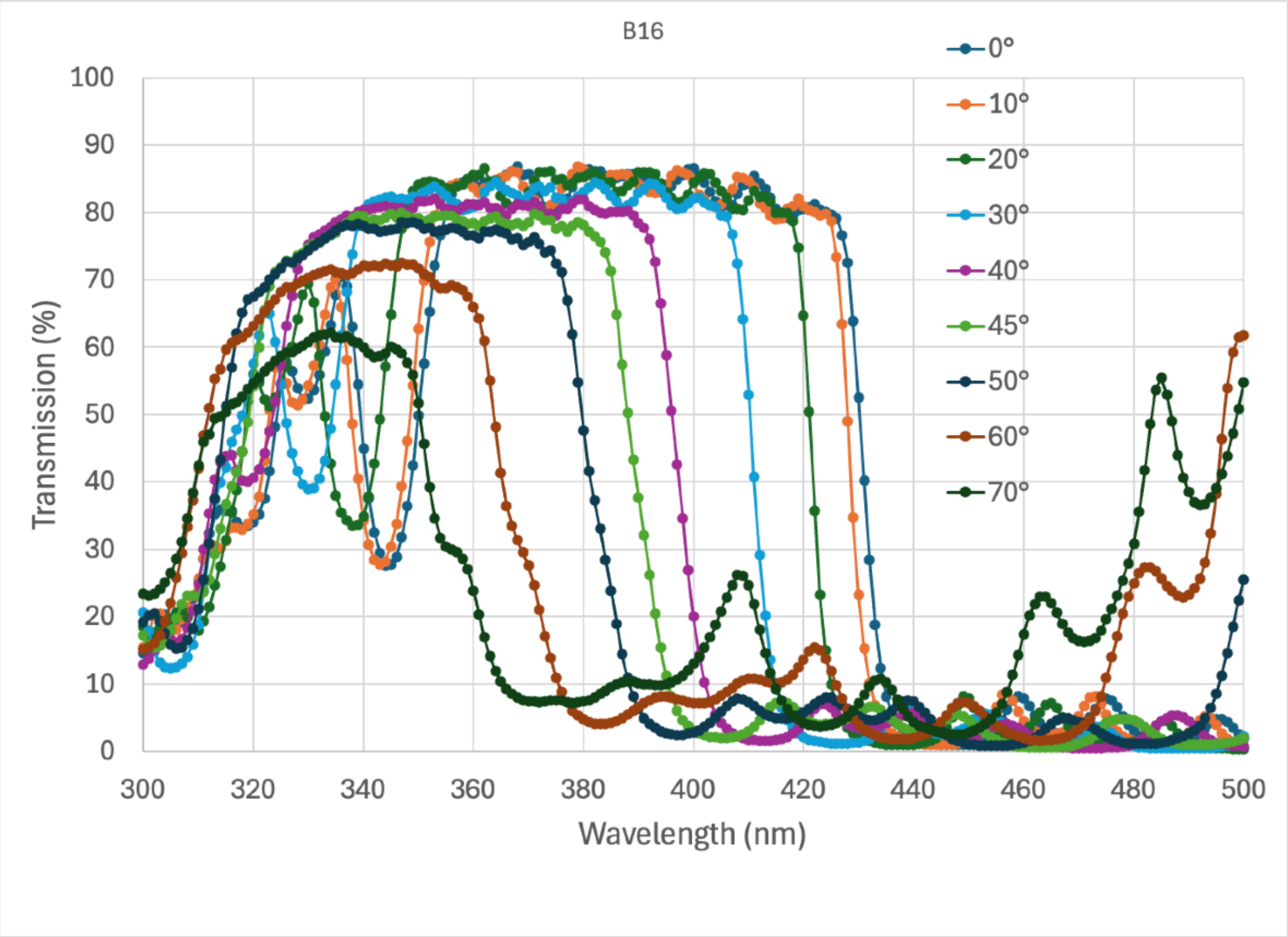
Sapphire sample B15 has coating HfO2/SiO2. Edge shift on both of these is larger.

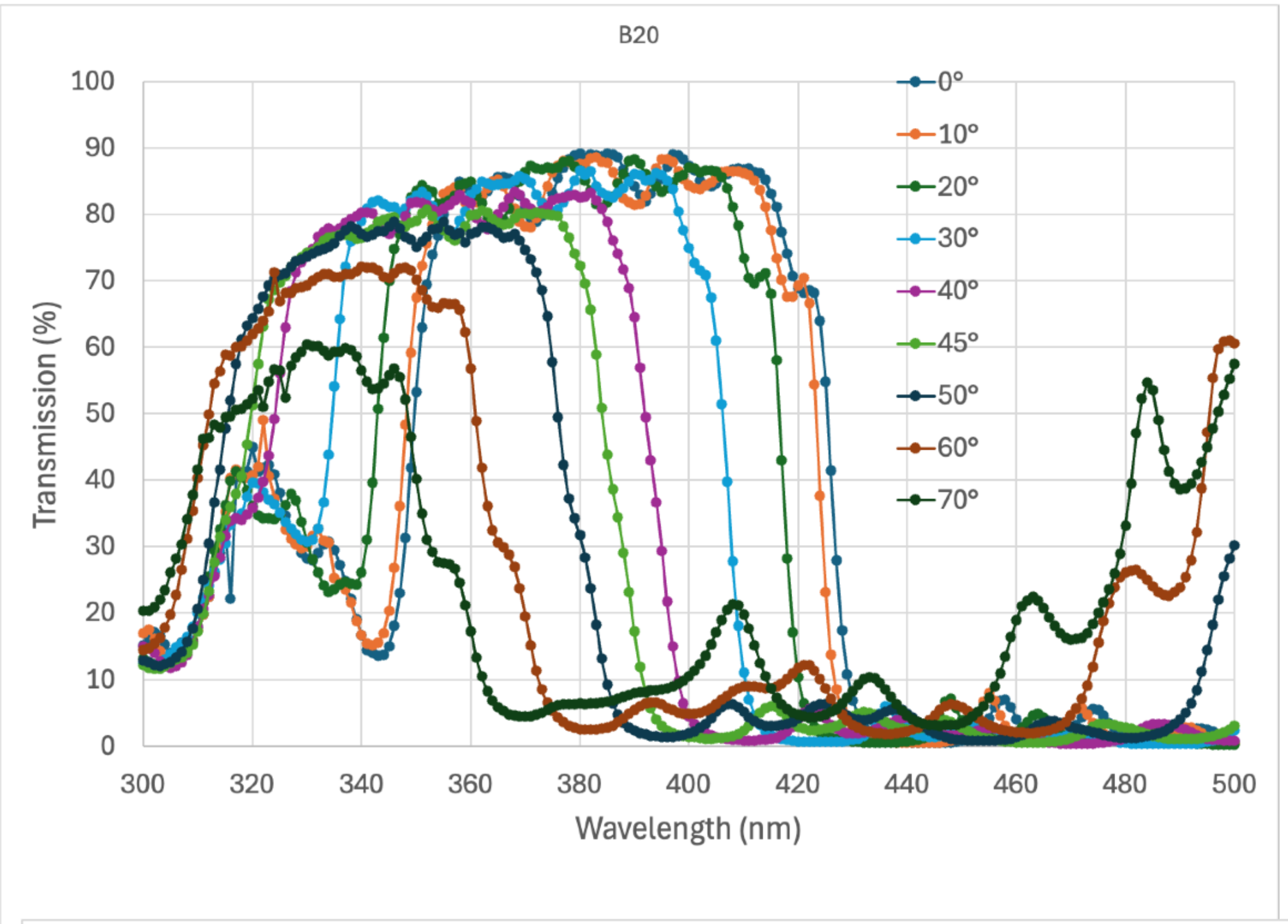
Key result: HFO2/Al2O3 with Fused Silica substrate will be the best

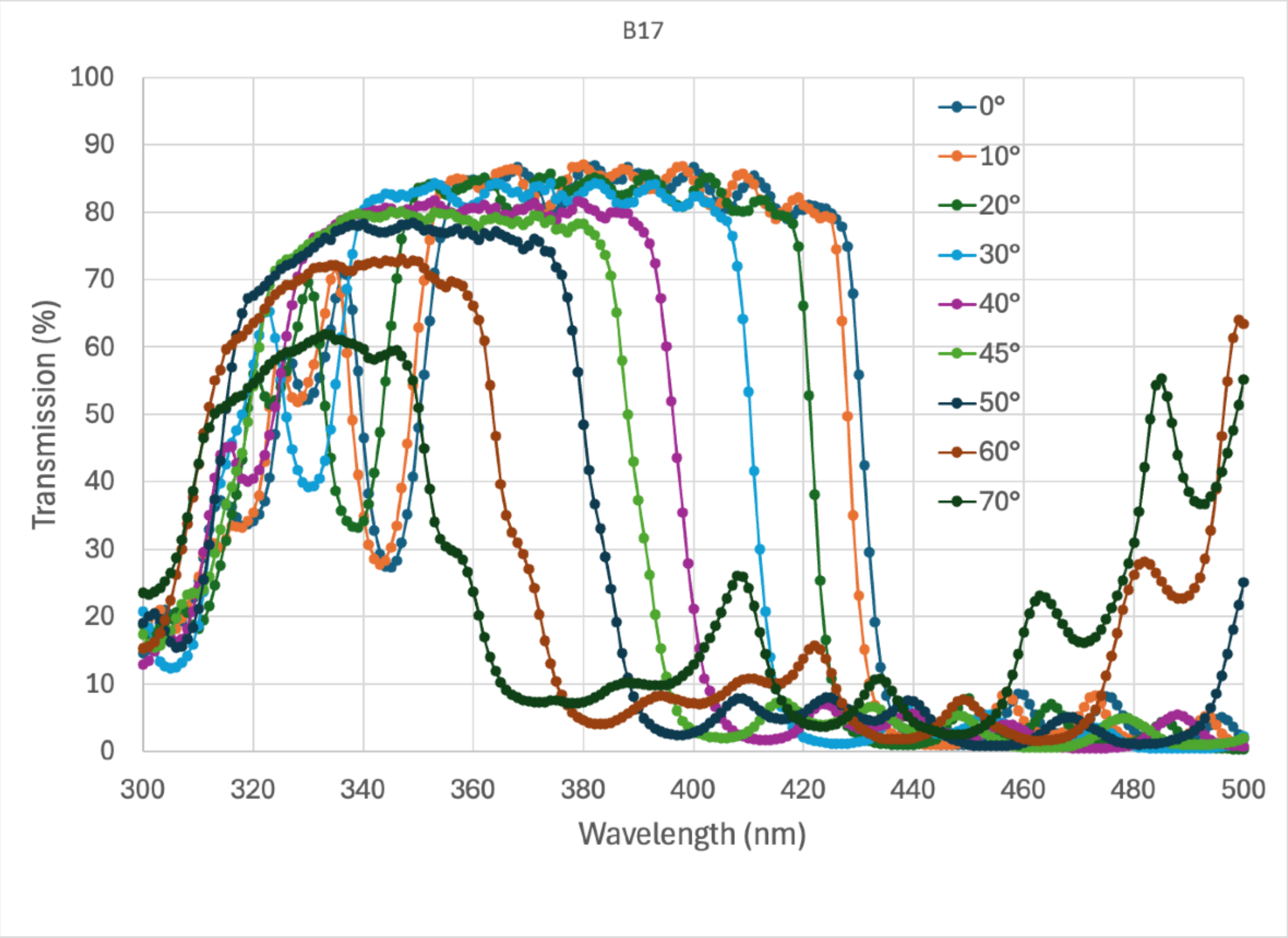
BNL Samples measurements

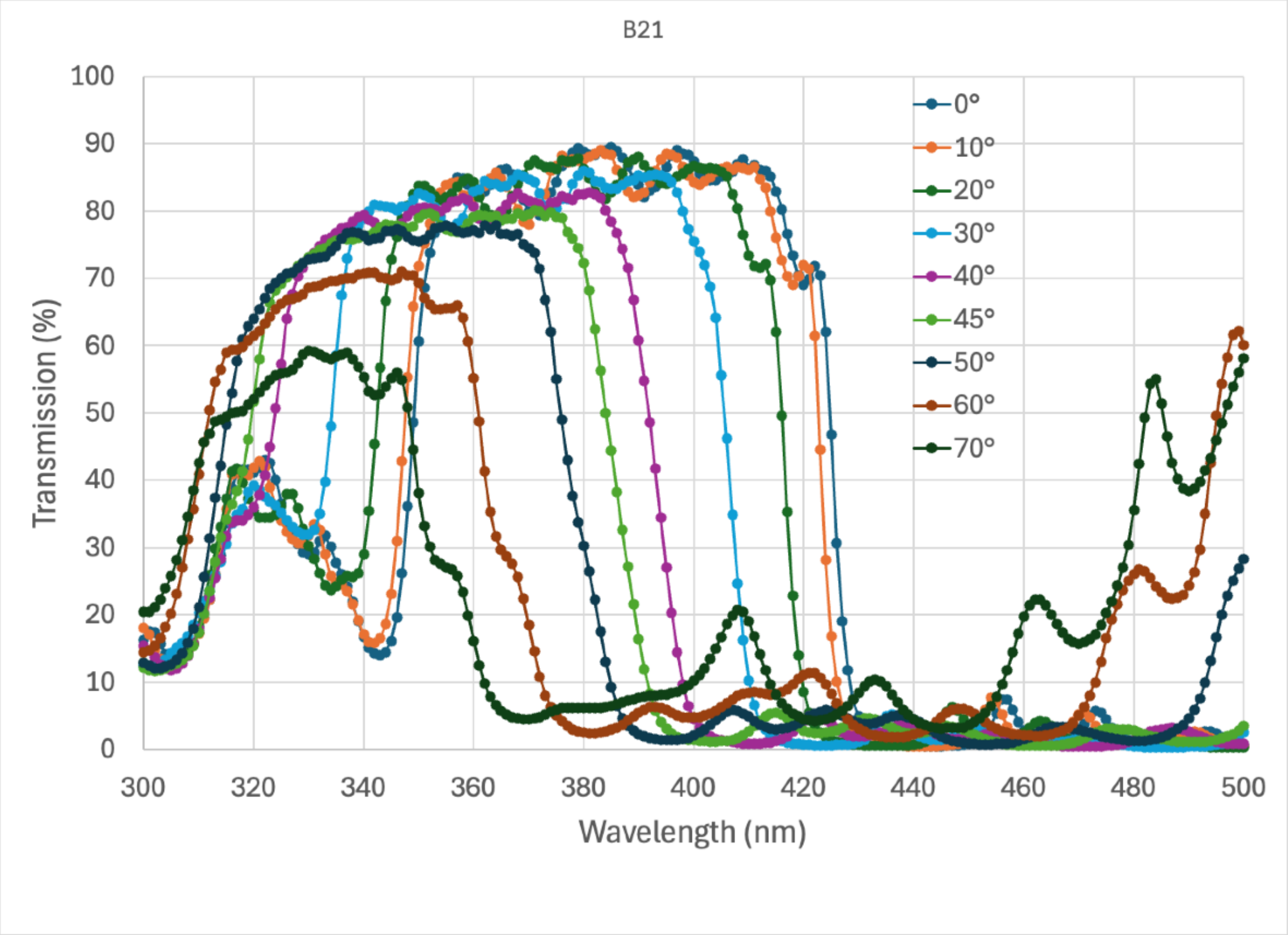
Sapphire

B33 glass









All need to be corrected for backside reflectance in transmission mode.

Transmission is close to 95% for 350 nm for sapphire, but B33 loses in the UV region.

These were made with the BENEQ machine.

Best angular performance is with the VECO200 produced filters.

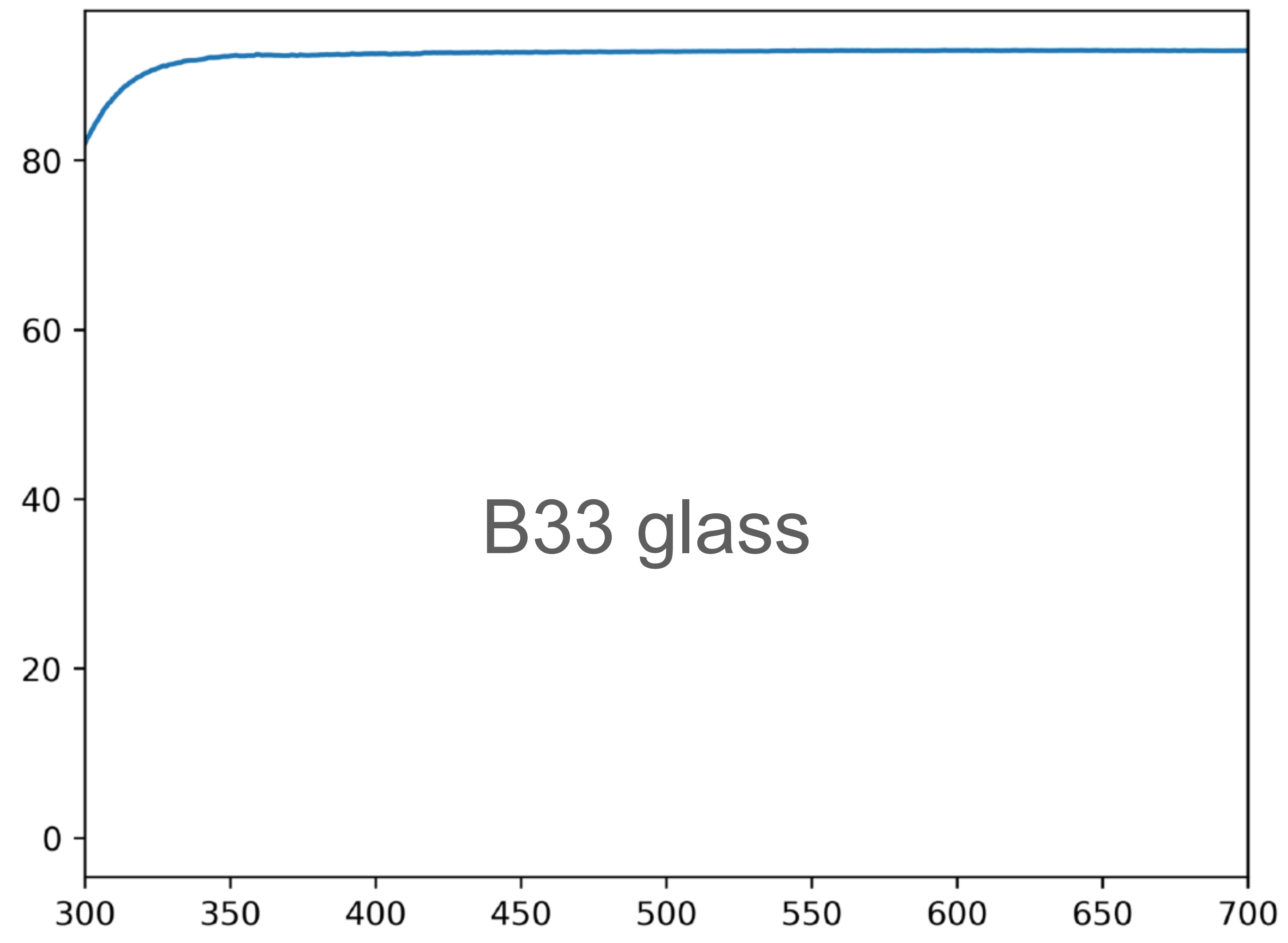
These measurements match well between BNL and RAYTUM.

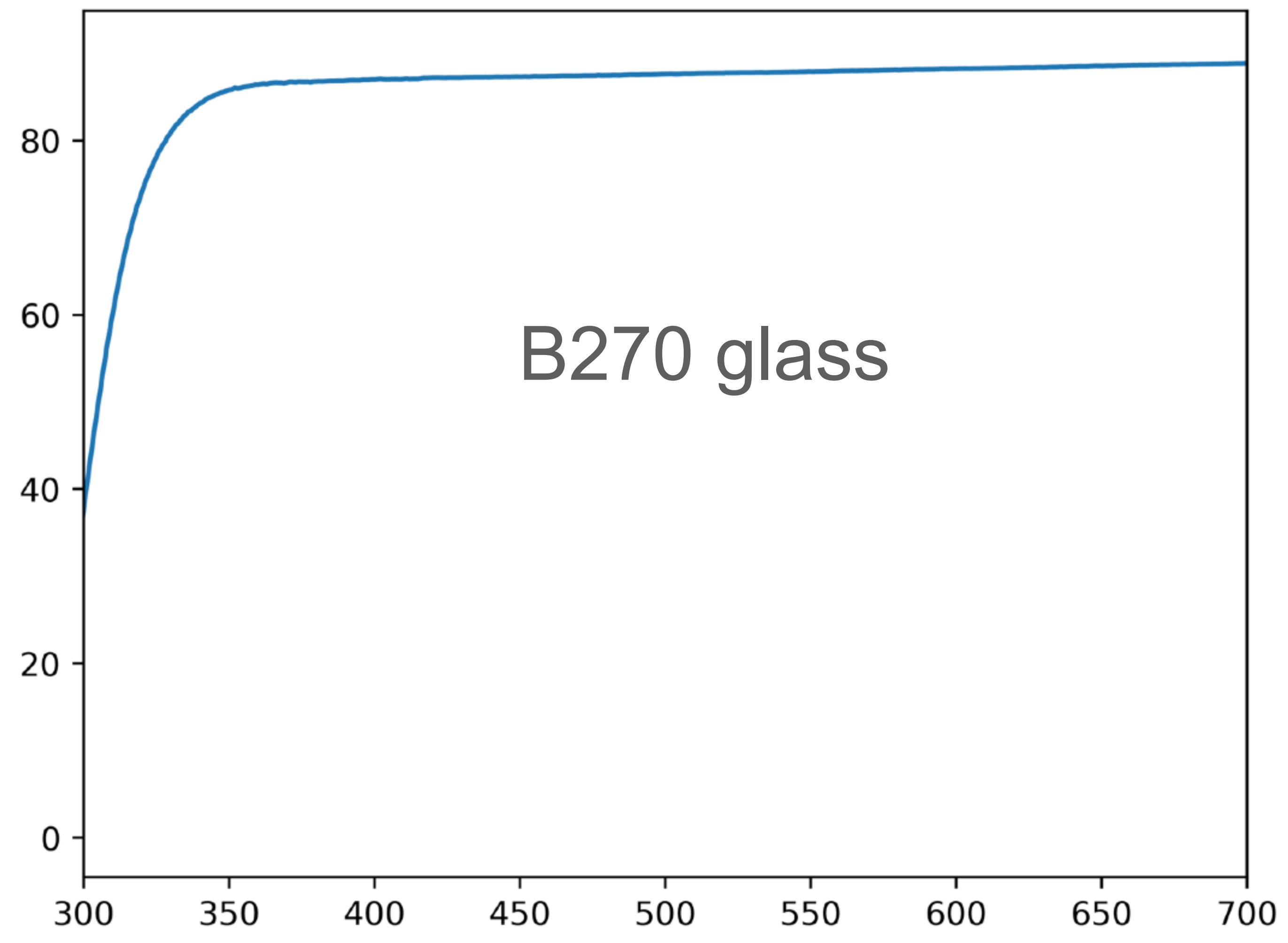
Conclusions from evaluation of ALD made large area SP DF performance

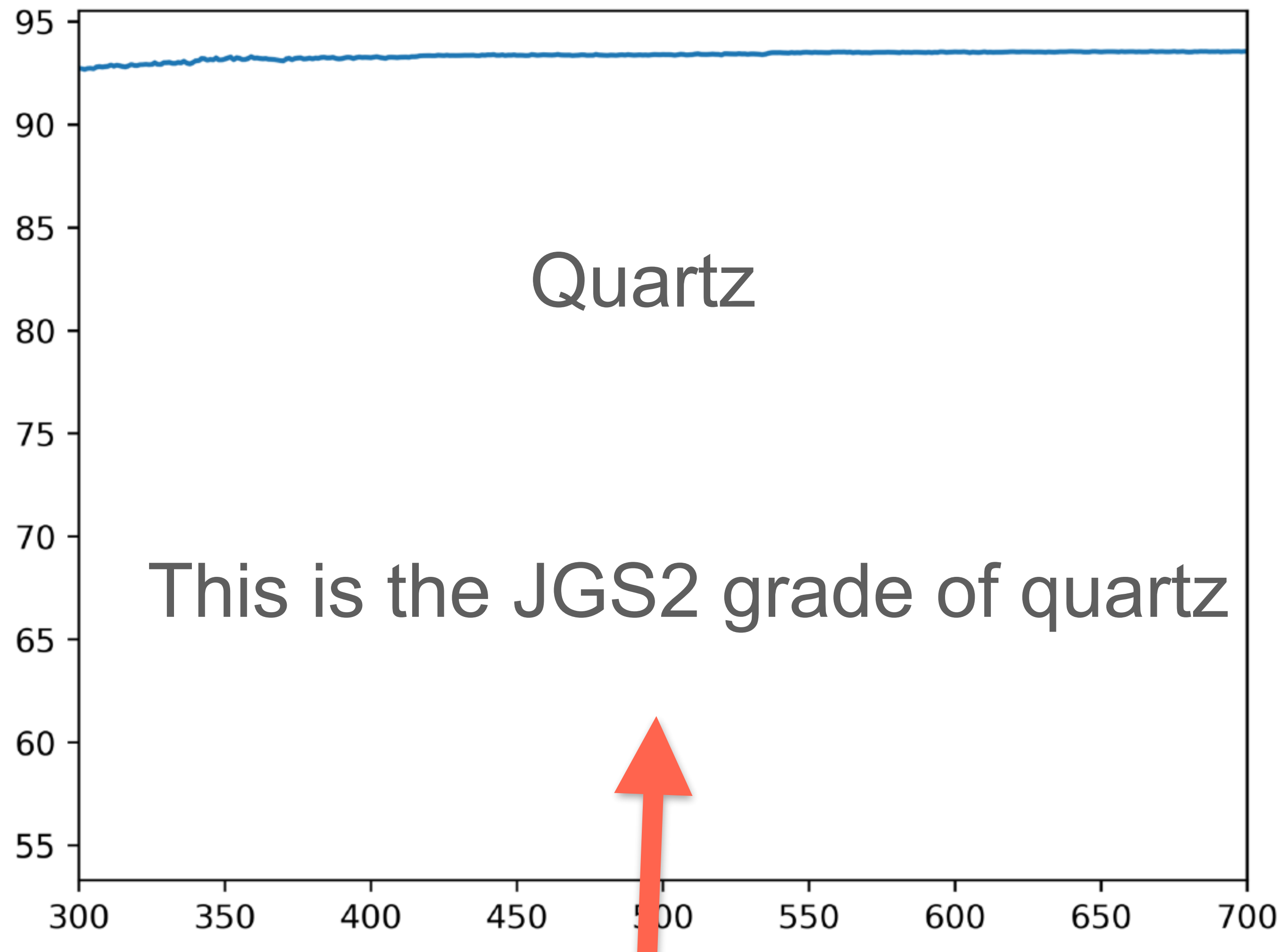
1. Short-pass dichroic filters consisting of either HfO2/Al2O3(HA) and HfO2/SiO2(HS) material system were designed and optimized at 45° in 320-500nm wavelength range with a cutting–off edge at 380-400nm.
2. Sapphire, SiO2, B270, B33 optical substrates with sizes in 4 inch diameter or 100mmx77mm square were used to optimize residual stress, UV transparency and cost effect.
3. SP DFs were fabricated by either Veeco 200 temporal ALD using thermal ALD mode or Beneq C2R spatial ALD in PEALD mode.
4. ALD process conditions, precursors used, optical absorption and scattering of single layers, stress data, etc.
5. Optical transmission and AOI dependence were evaluated extensively and independently by ARL, RP, Beneq and UPenn using commercial UV/Vis spectrometers or RP home setup.
6. Both HA and HS filters show excellent transmission > 95% in the transparent band(<380-400nm) and low transmission(5-10<%) in the blocking band(380-500nm) if backside reflectance from different substrates are subtrated. A std broadband AR coating(320-380nm) may be added on back side in future to minimize substrate effects. Of course a good filter design should be optimized at least on top surface of different substrates with different index.
7. Cutting-edge broadening <10nm.
8. AOI dependence from 0-70° in air show a narrow edge shift of 45nm measured at 50% cutting-off edge transmission.

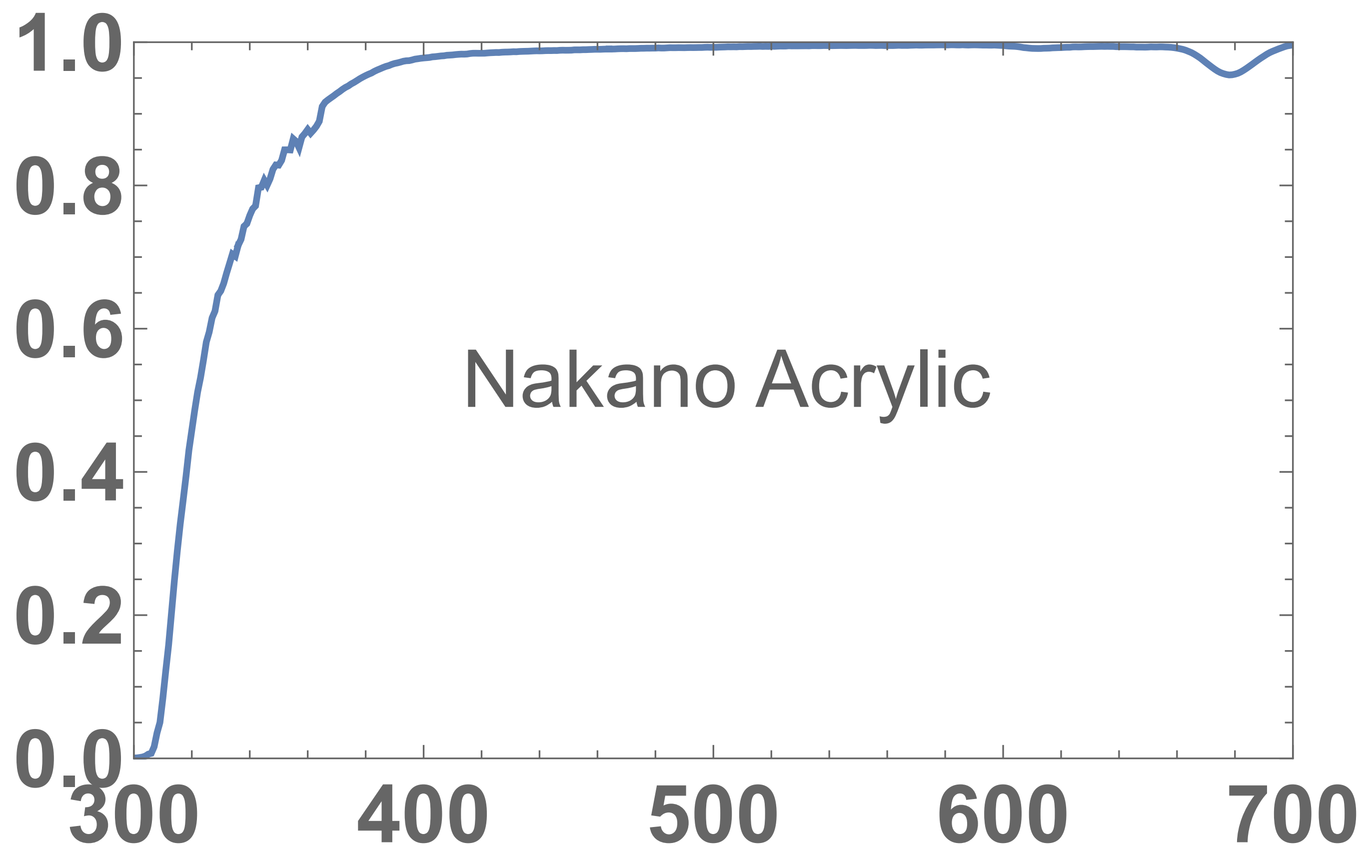
Information on Substrates

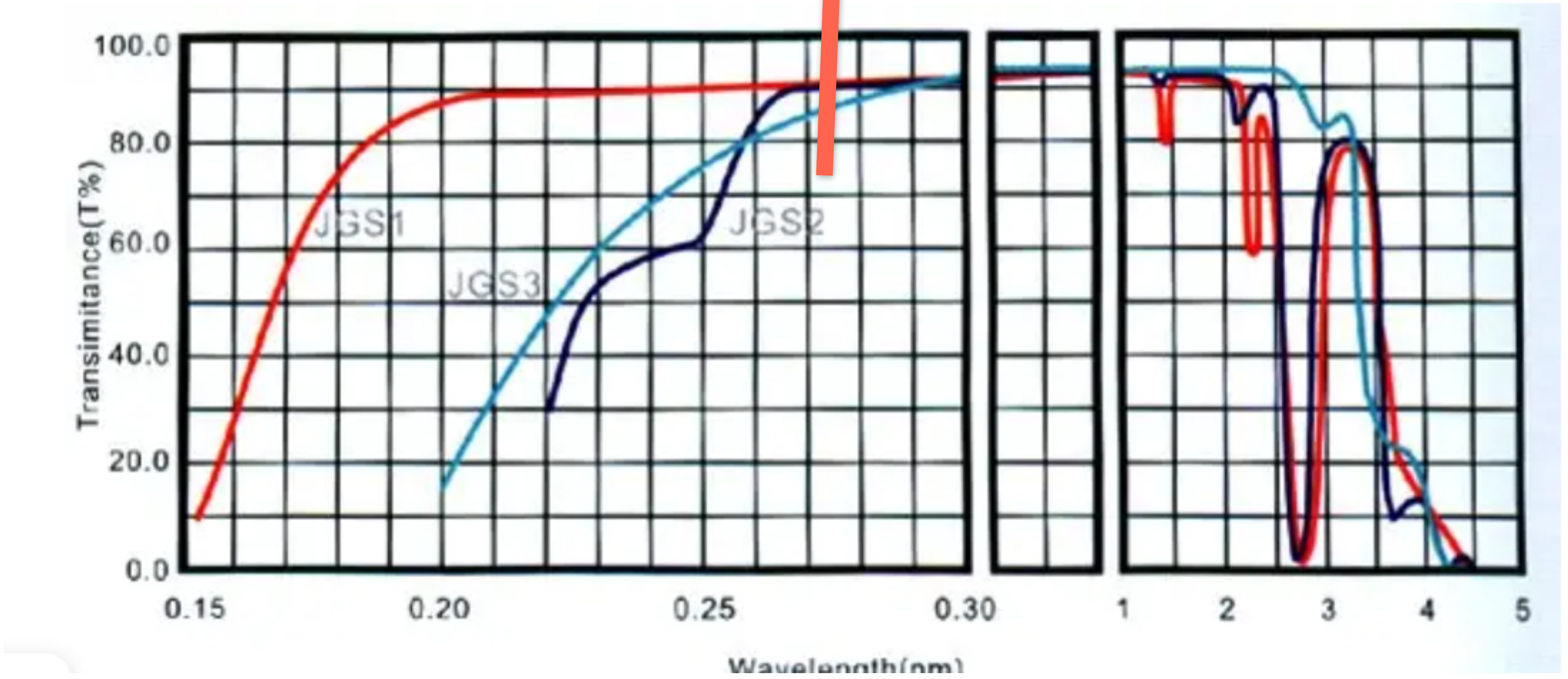
Available: Sapphire, B33 glass, B270 glass, Fused silica, Best acrylic











- Transmission was measured in air for 1 mm samples at BNL. Add back the reflectance from both surfaces ~ 8 % (except for acrylic which is already corrected)
- Sapphire is excellent down to 200 nm, but expensive.

Next steps in progress with DUNE

DUNE FD2 size filters have been ordered

Table 1

Task	Dimension	Substrate	Dichroic side			comment	plan
Samples for measurements	77.0 +- 0.3 mm	Sapphire	One side coated 380 mn cu	Beneq	2	BNL measured	Brazil for PTP coating
	100.00 +- 0.3 mm	B33	One side coated 380 nm cu	Beneq	2	BNL measured	Brazil for PTP coating
	1.00 +- 0.1 mm	Sapphire	One side coated	Raytum	3	Raytum measured	Brazil for PTP coating
		B270	One side coated	Raytum	1	Raytum Measured (probably not suitable.	Brazil for PTP coating
		Fused Silica	One side coated	Beneq	4	Not yet ready	Brazil for PTP coating
Prototypes for DUNE-FD2	143.75+-0.3 mm	Sapphire	One side coated 380 nm	Beneq	20	Purchased	See comment on PTP coating
	143.75+-0.3 mm						Do you want an adhesion layer ?
	1.5 +- 0.1 mm	B33	One side coated 380 nm	Beneq	20	Are these needed ?	
		Fused Silica	One side coated 380 nm	Beneq	40	Purchased	
						Substrate cost	
Estimate for DUNE -FD2 productio	143.75+-0.3 mm	Sapphire	One side coated 380 nm		17000	$74.70/each	
	143.75+-0.3 mm						
	1.5 +- 0.1 mm	Fused silica	One side coated 380 nm		17000	$11.60/each	
		B33	One side coated 380 nm		17000	$3/each	

We buy B33 and B270 from this supplier: https://www.howardglass.com/spec-sheets/

Additional Comments and PTP coating

Options need to be explored

The filter on quartz can only be coated by the Beneq machine, whose stress-free plasma process can match the low CTE of quartz.   I will keep you posted when the new batch of filter on Quartz is scheduled.

PTP coating on various substrates could be an R&D topic of importance. There are two ways to go

1) An adhesion layer with ALD can be deposited for the PTP coated side. This can be minimum cost.
2) A low temperature plasma system can be used to energize the substrate surface before PTP coating.

Typical Kurt Lesker Nano 36 Thermal Evaporator Thin Film Deposition

$20000

Questions that need to be answered in collaboration with DUNE

- A lot of information on substrates and possibilities for coatings is in hand.
- FD1 and FD2 may not be using dichroic filters.
- Nevertheless, some units can be supplied for a final test. These will be with fused silica without PTP coating.
- Can we narrow options for FD3/FD4 ?
  - What substrate and size would you like ?
    - BENEQ can deposit up to 50 cm x 50 cm
  - What is the requirement for the angle ? What is the movement of the cut wavelength with angle. What options for the wavelength shifter coating.

Option	**Substrate**	**size**	**with dichroic at 380 nm**	**PTP coating**
1	B33	~ 50 cm	Yes/No	Done by industry with adhesion treatment
2	fused silica	~ 50 cm	Yes/No	same
3	fused silica	~ 50 cm	Yes/No	No PTP coating
4	Acrylic	~ 50 cm	Needs work	with PTP coating

Thermal Shock test of ALD coated Filters on 77mmx100mm substrates

Tests are successful on B270, B33 and Sapphire substrates as long as cooling is reasonably slow.

Four tests are performed in sequence:

Test 1: A piece of SiO_2/HfO_2 SP filter on B270 substrate, diced from the original 77mmX100m filter, was tossed directly into liquid nitrogen and take out and let warm up in atmosphere after reach thermal equilibrium (stop bubbling).

Test Result: The B270 glass shattered upon touching the liquid nitrogen, but the thin film on the remaining substrate is intact.

Test 2: test 1 is repeated with the same type of SP filter on B270 substate sealed in a plastic bag

Test Result: The filter is intact. There is no visible changes on the filter

Test 3: test 1 is repeated with the same type of SP filter on B33 substate sealed in a plastic bag

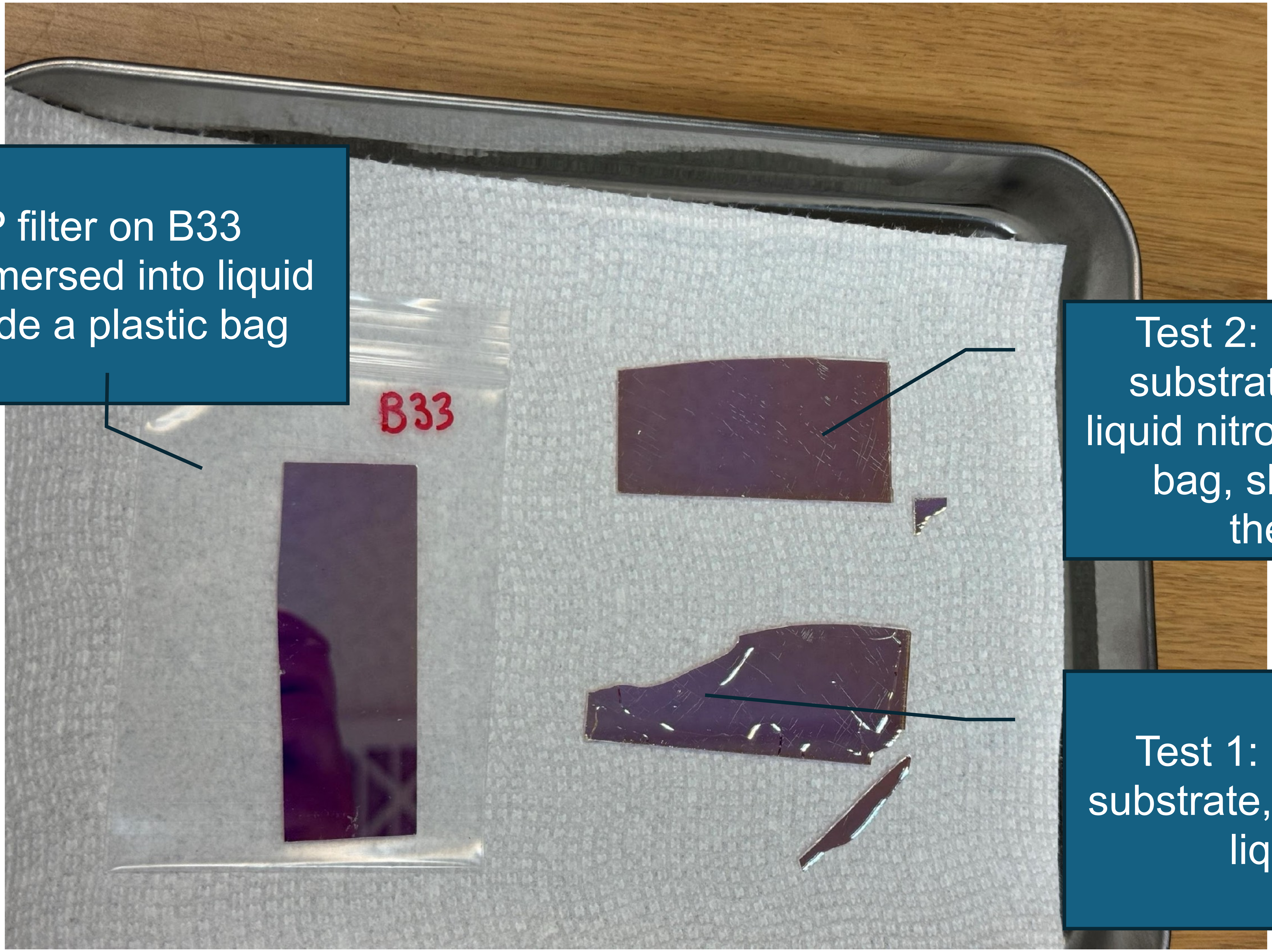
Test Result: The filter is intact. There is no visible changes on the filter

Test 4: test 1 is repeated on the same type of SP filter on sapphire substate sealed in a plastic bag. The sapphire filter is an entire 77mmx100mm

Test Result: The filter is intact. There is no visible changes on the filter

Video of the experiment

Photos taken after immersed into liquid nitrogen.

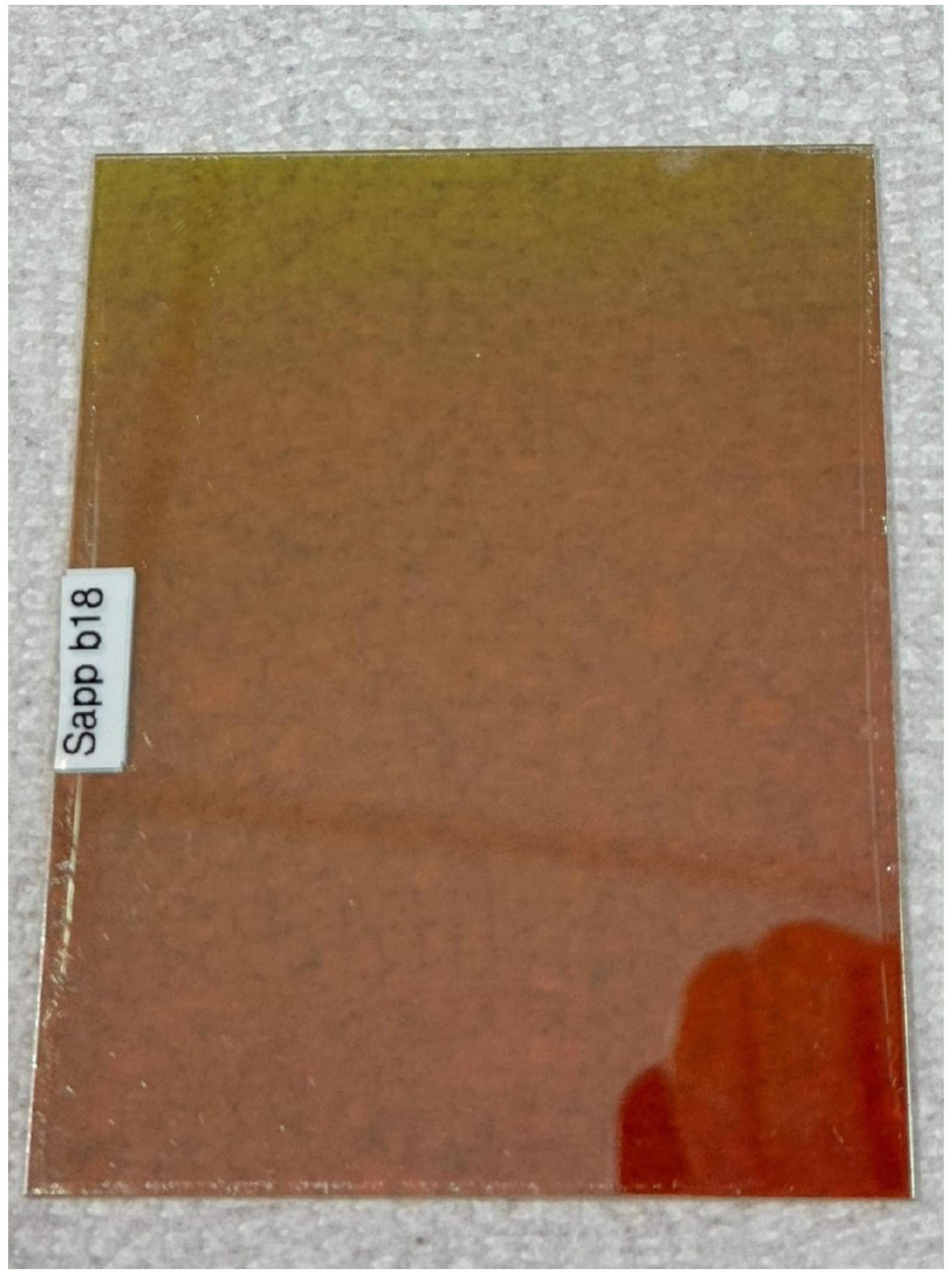


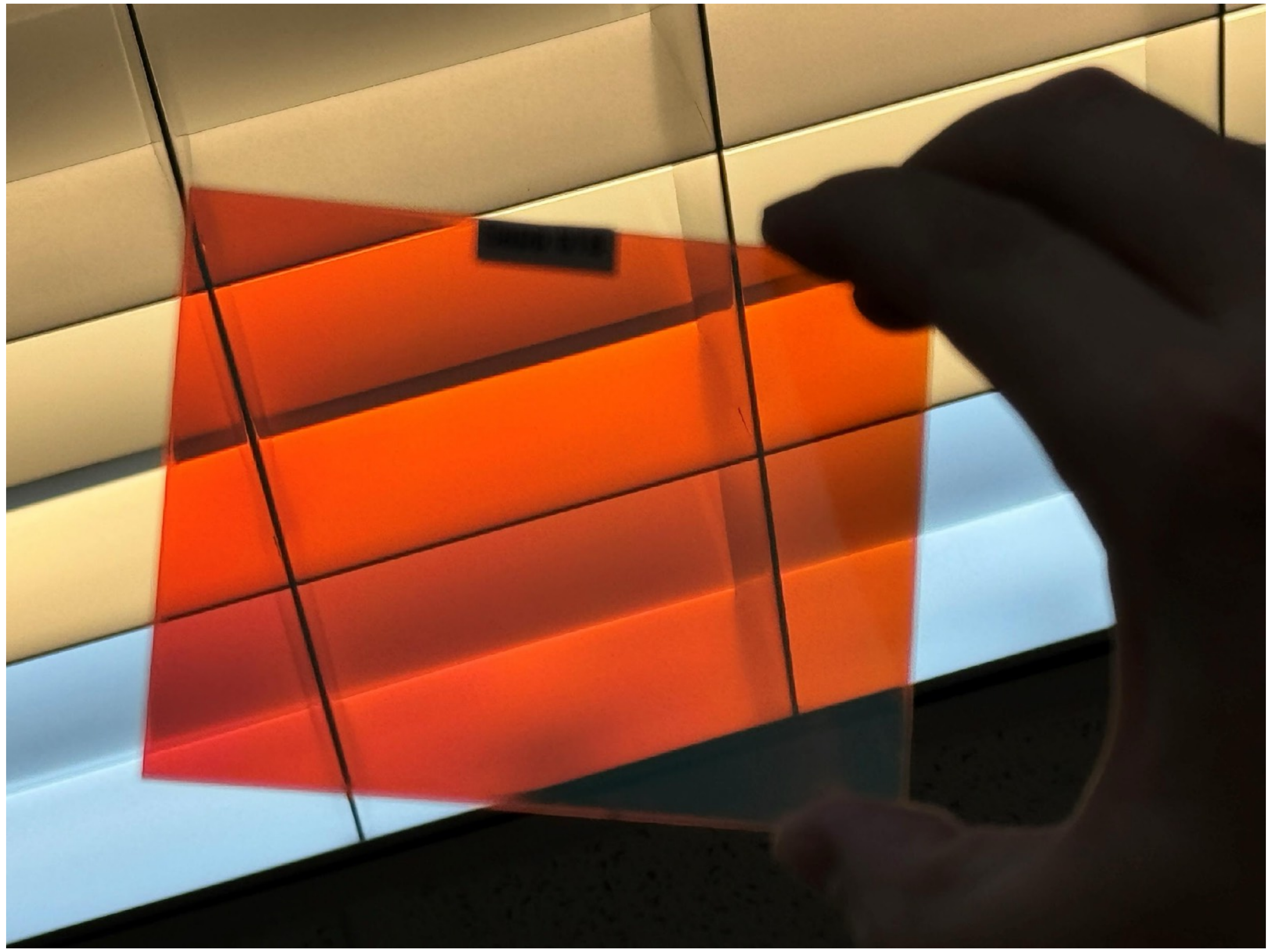
Test 3: SP filter on B33 substrate, Immersed into liquid nitrogen inside a plastic bag

Test 2: SP filter on B270 substrate, Immersed into liquid nitrogen inside a plastic bag, slowing down the thermal shock

Test 1: SP filter on B270 substrate, directly tossed into liquid nitrogen

Photos taken after immersed into liquid nitrogen. A 77mmX100mm SP filter on Sapphire substrate is intact (test 4).



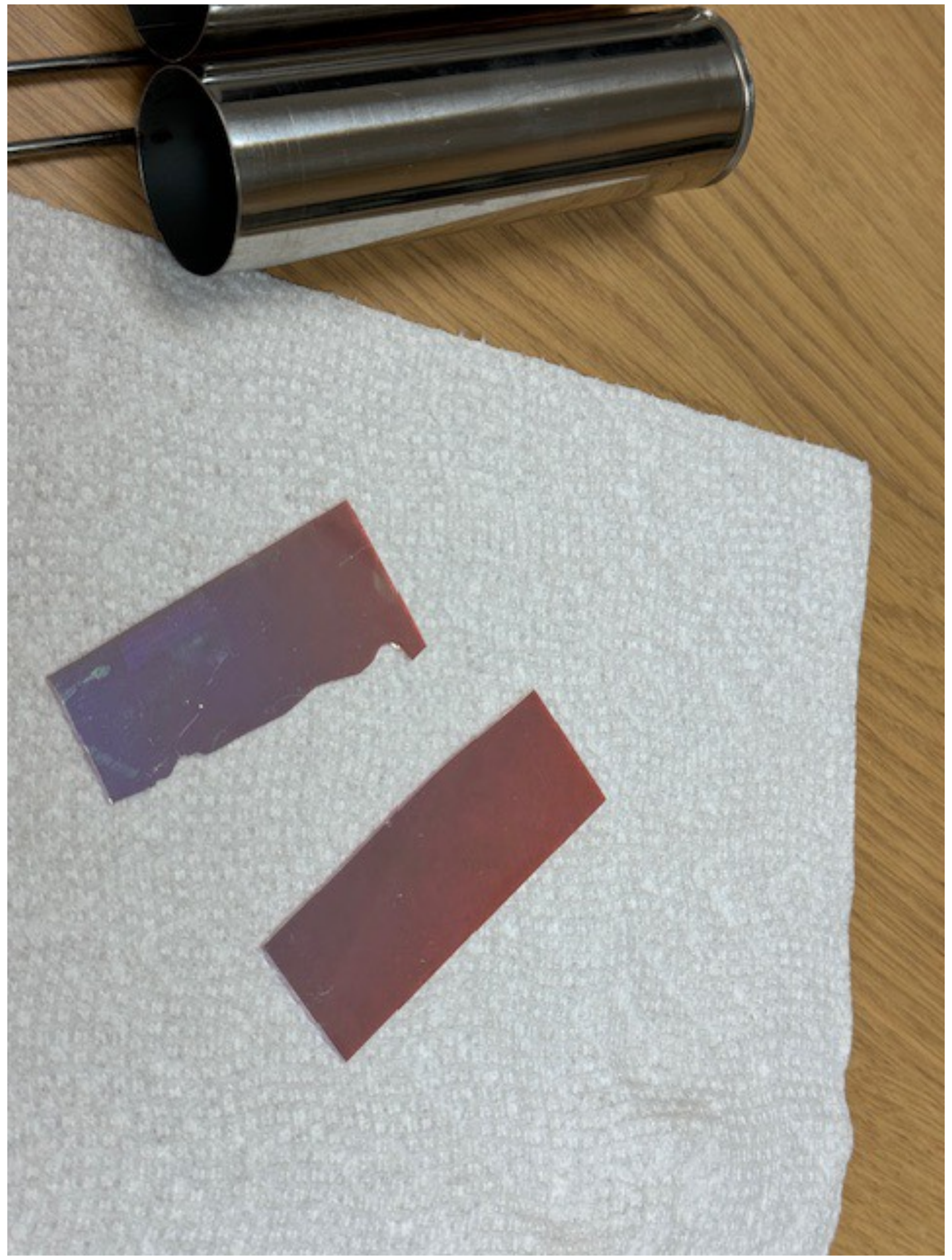


Longer term test

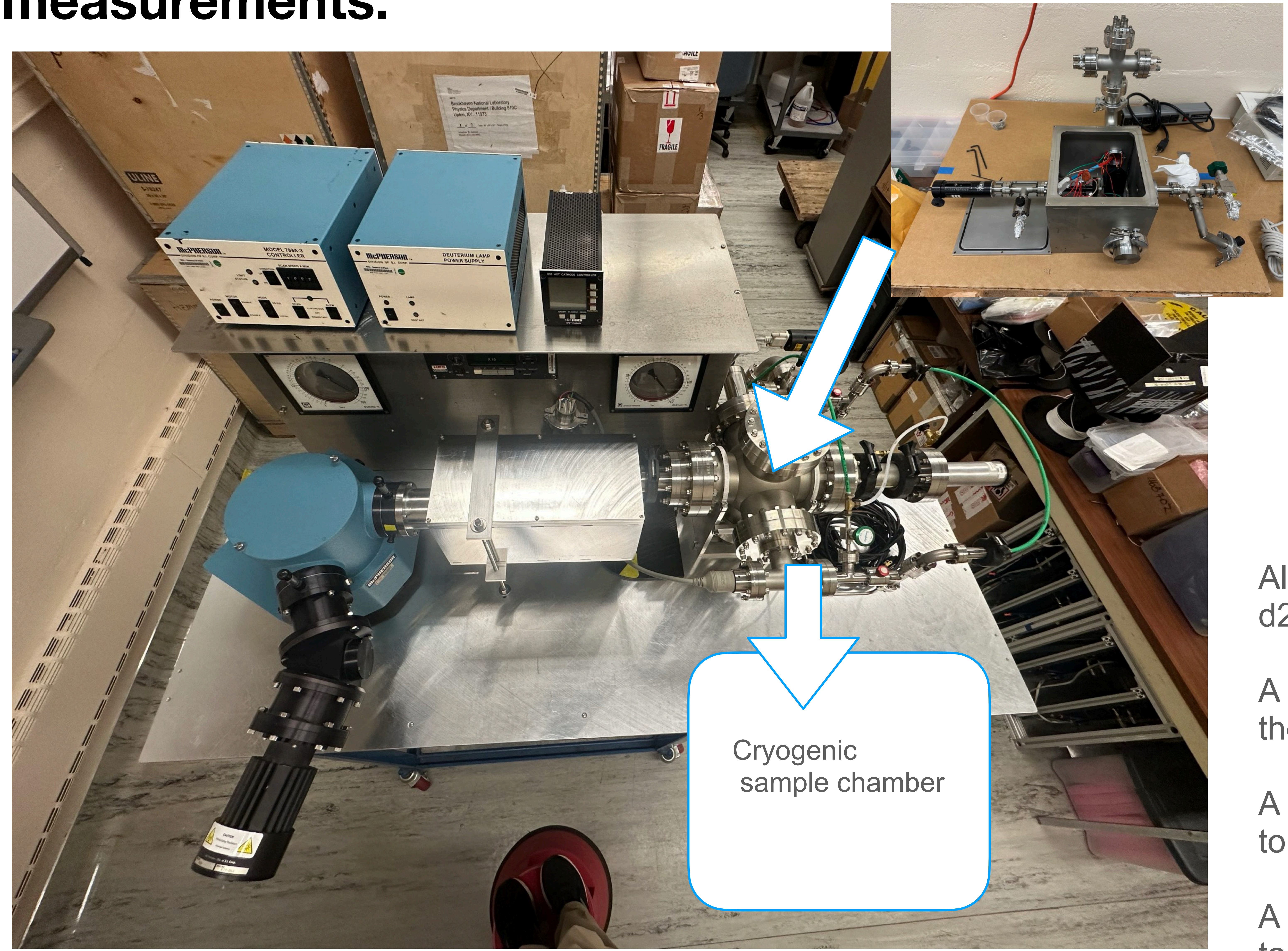
In LN2

2 pieces of SP filters, one on B270 glass and another one on B33 glass, were immersed in liquid nitrogen continuously for 15 days.

Both filters are intact after returning to atmosphere.



BNL is building a specialized photo-spectrometer for absolute sensitivity measurements.



All new optics will be installed with very bright d2 lamp.

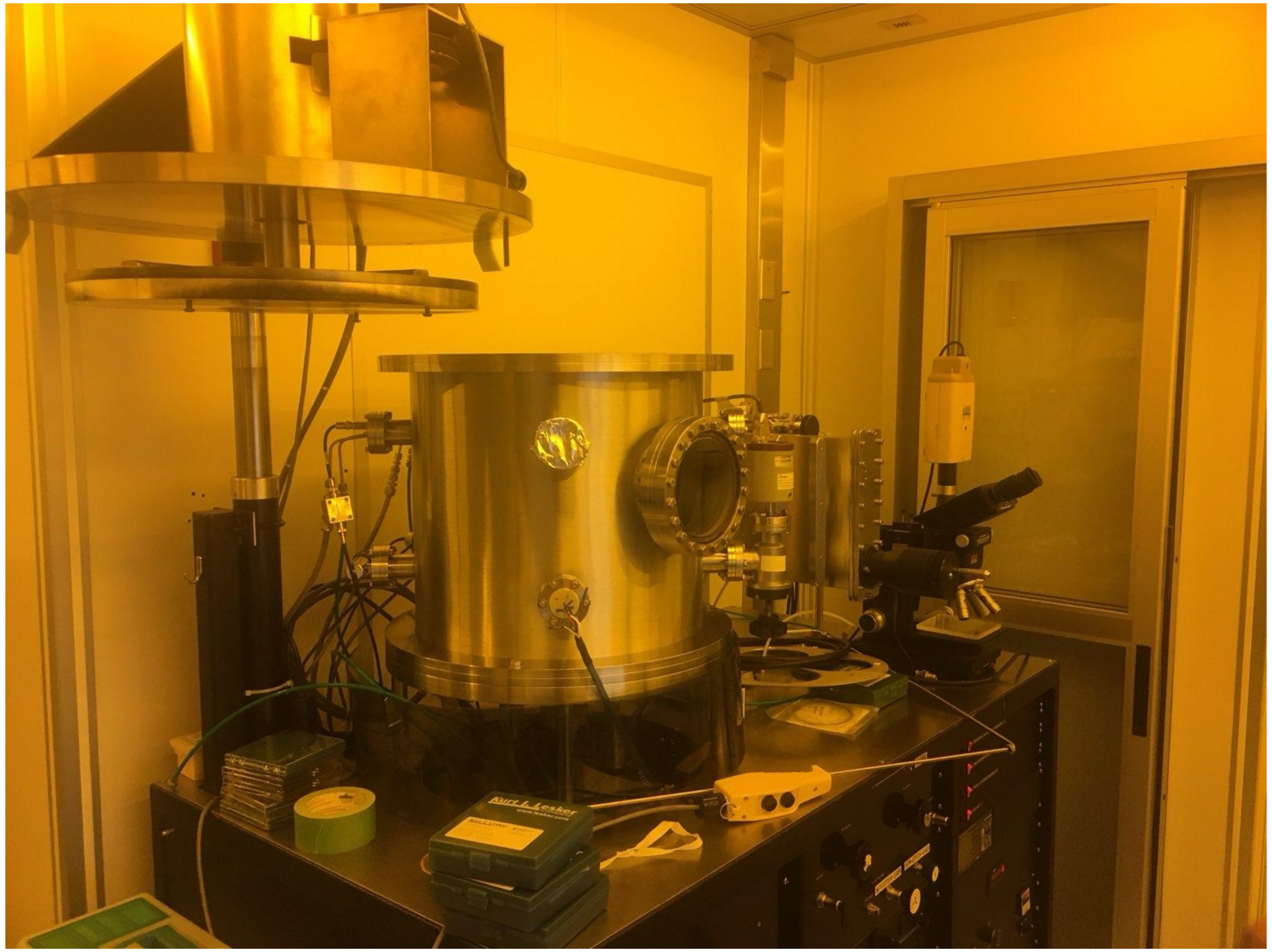
A NIST calibrated set of PMTs will calibrate the beam from 110 to 600 nm.

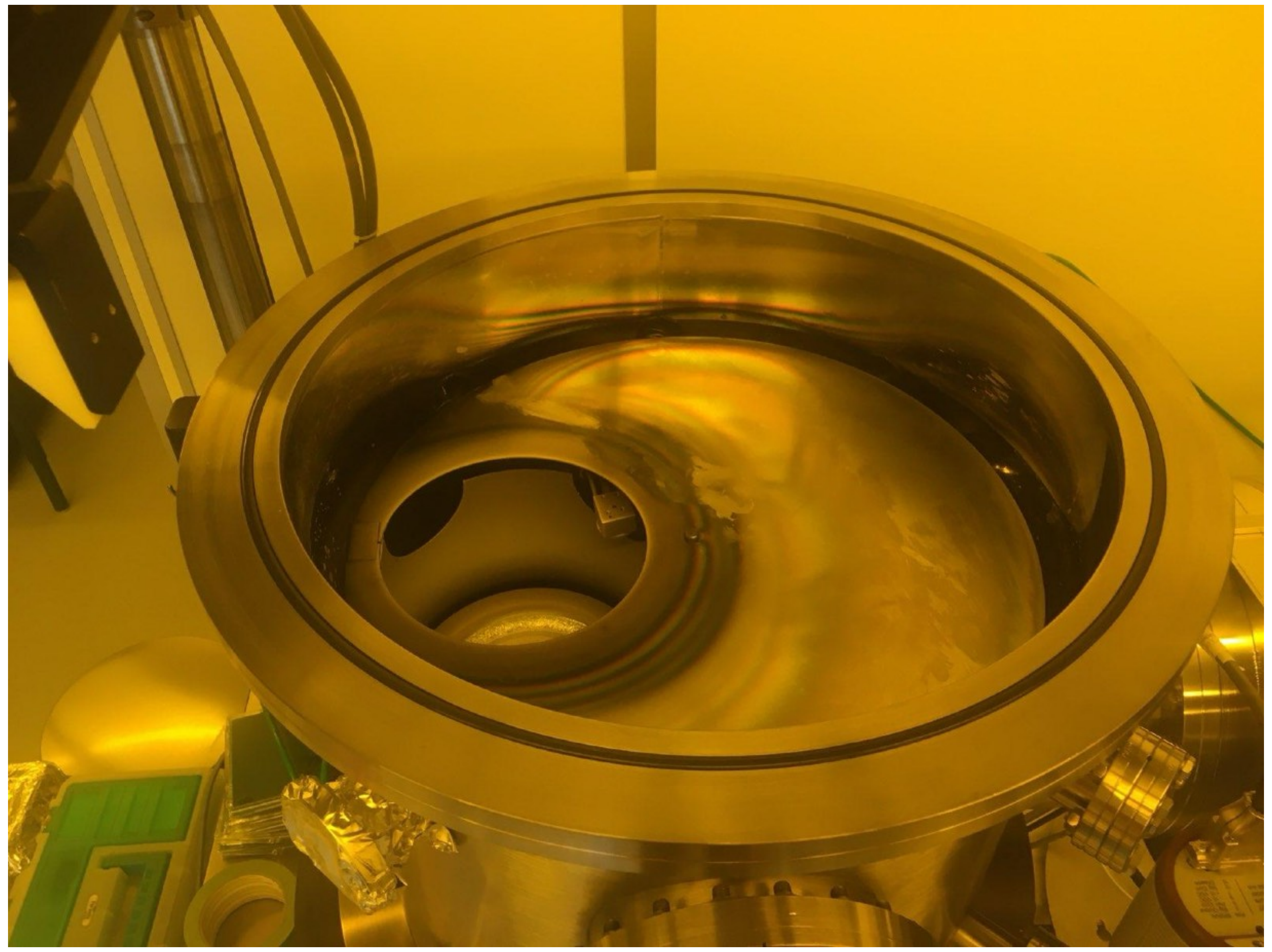
A beam chopper will be added to send beam to calibration and to sample

A sample chamber will be added below the table for large devices.

BNL facilities for PTP, TPB coatings

There are many idle vacuum deposition machines in US national labs. They could be dedicated to single molecule.





Specifications

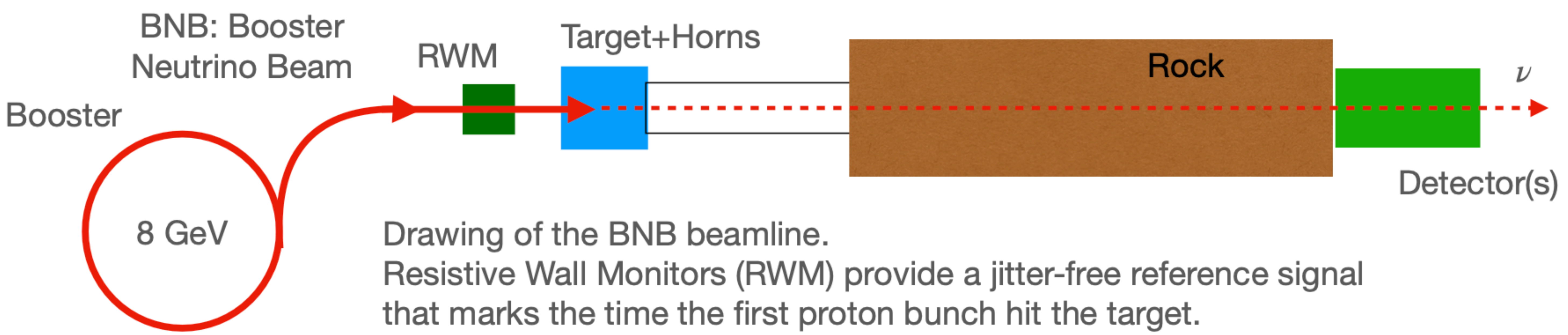
~60 cm inner ID.

~needs small investment to make it is compatible with PTP and dedicated 30-60 minutes for batch of 10 possible.

Conclusion

- A project using atomic layer deposition ALD for carefully tuned high quality dichroic filters will complete in September 2024.
- An important break-through in achieving high transmission from high index layers has been achieved.
- Design of filters needed for DUNE or THEIA has been demonstrated.
  - A lot of data can be made available for optimization.
- Preproduction prototypes have been delivered.
- Further collaboration on testing is very much needed.
- A next phase of SBIR is possible, but needs close collaboration at this stage. A lot of flexibility is possible in coatings and optimization.
- ***A caution: The optical performance is quite complicated and needs a proper simulation.***
- ***For further work it is best to work directly with this company.***

Neutrino beam timing

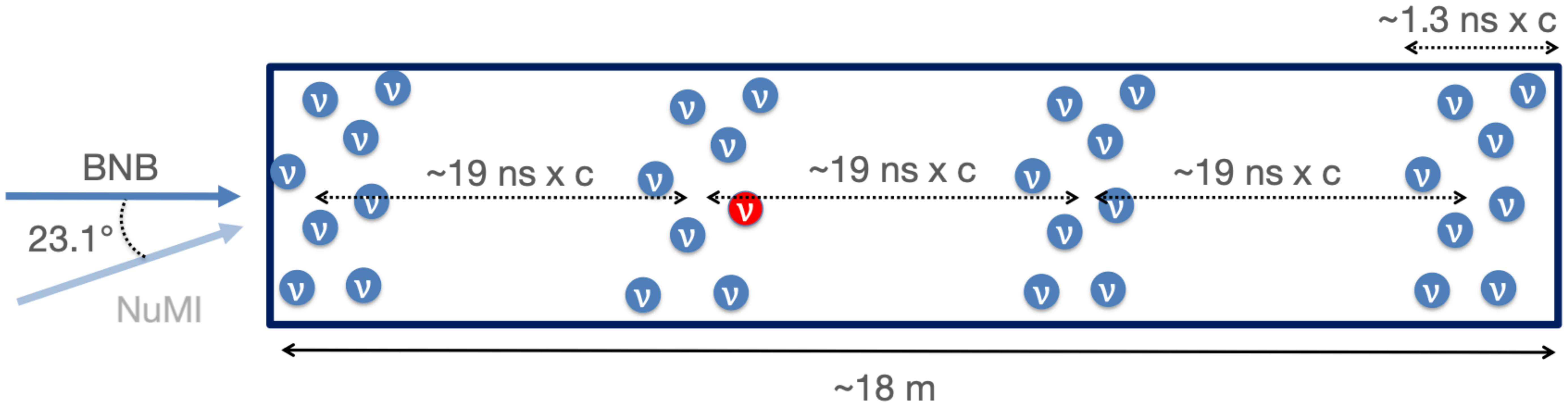


Drawing of the BNB beamline.
Resistive Wall Monitors (RWM) provide a jitter-free reference signal that marks the time the first proton bunch hit the target.

Beam timing:

- BNB: 1.6 μs spill, 81 bunches, 18.9 ns spacing (52.8 MHz)
- NuMI: 9.6 μs spill, 486 bunches, 18.8 ns spacing (53.1 MHz)

Neutrino propagation (+ meson decays) only adds a constant offset, so neutrinos inherit the **time profile of the proton bunches.** Precise event timing allows to tag neutrinos directly with no use of charge!



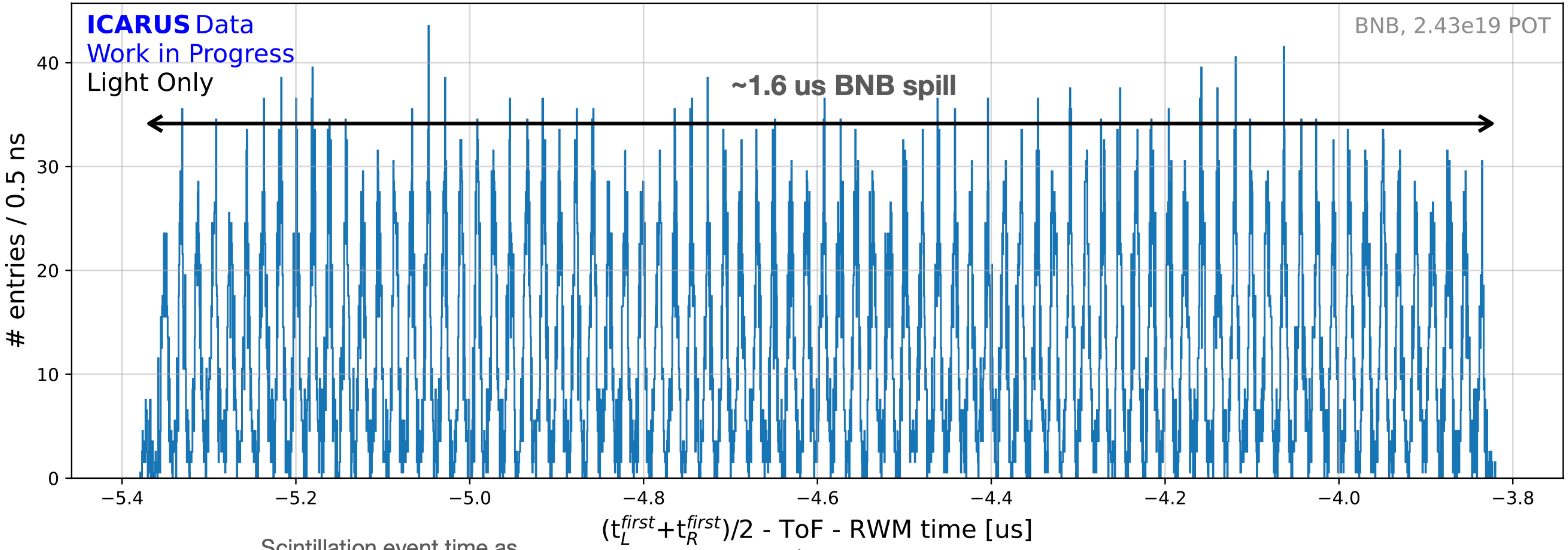
BNB bunches crossing ICARUS

Achieving this goal requires a relative and absolute calibration strategy towards **O(ns)** resolution!

BNB bunch structure

Light-only reconstruction

(After subtracting cosmics)

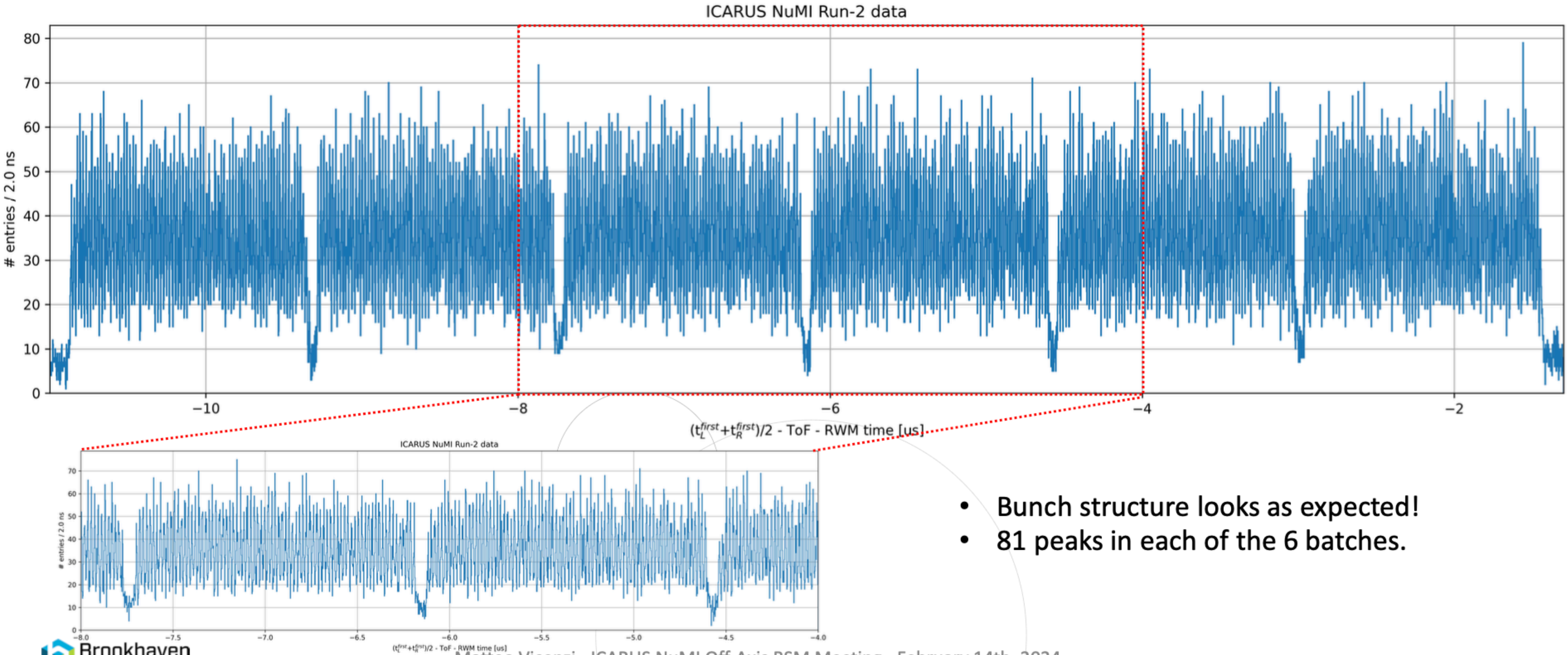


Scintillation event time as mean between first PMT times on opposite walls

Time-of-flight correction from light barycenter

Synchronous to accelerator cycle

NuMI bunch structure



- Bunch structure looks as expected!
- 81 peaks in each of the 6 batches.