

Controlling the Stability of Xenon-Doped Argon Mixtures

Ethan Bernard

Eli Mizrachi, Jimmy Kingston, Jingke Xu, Sergey Pereverzev, Teal Pershing, Ryan Smith, Charlie Prior, Nathaniel Bowden, Adam Bernstein, Carter Hall, Emilia Pantic, Mani Tripathi, Dan McKinsey, Phil Barbeau

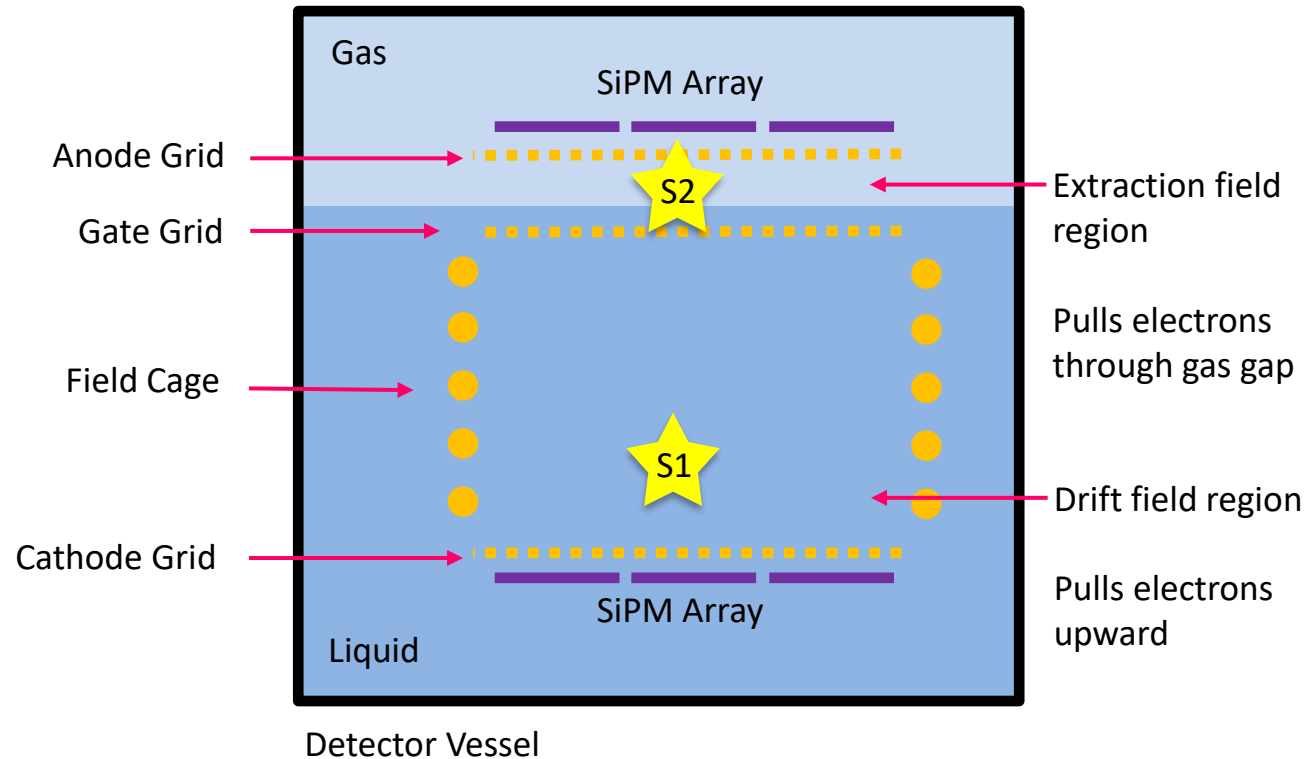
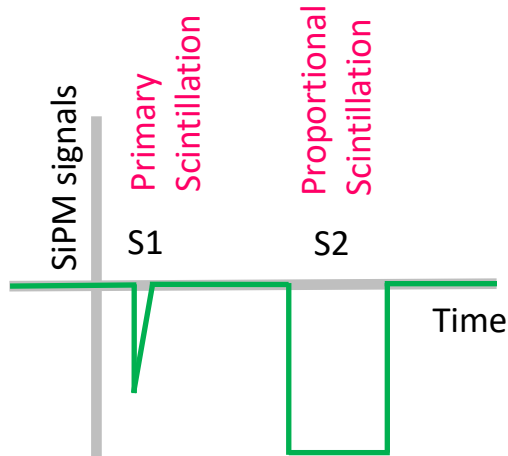
LLNL,
University of Maryland,
UC Davis,
UC Berkeley

arXiv: 2209.05435

CPAD 2022
November 29th, 2022



Dual Phase Noble Liquid TPC Operation



Pure Ar: *Few μs*
Metastable Gas dimer \rightarrow 128 nm photons \rightarrow Excited TPB molecules \rightarrow 420 nm photons \rightarrow Photodetectors

Pure Xe: *Tens of ns*
Metastable Gas dimer \rightarrow 178 nm photons \rightarrow Photodetectors

Detection Medium Properties

- Single-phase liquid argon is the workhorse target medium for low cross-section physics
 - ICARUS T600, MicroBoone, DUNE, and many others*
 - Most experiments sense charge electronically, not through the more sensitive electroluminescence mechanism
- For detecting the lowest energy events, dual-phase xenon is the most successful medium
 - Electroluminescence mechanism allows resolution of *single drift electrons*
 - Nuclear recoils yield measured to 300 eV **
 - Electronic recoils resolved down to 186 eV ***
- Argon electroluminescence light is more difficult to produce and more difficult to sense, but argon is otherwise a more convenient material than xenon.

Property	Gas scintillation wavelength	Gas scintillation lifetime	Liquid phase ionization energy	Ease of purification	Cost	Kinetic match to light particles
Argon	128 nm	~ 3.2 μ s	14.3 eV	Easier	Cheap	A = 39.95
Xenon	178 nm	~ 22 ns	9.28 eV	Difficult	Expensive	A = 131.29

* K. Majumdar, K. Mavrokoridis, arXiv:2103.06395

** B.G. Lenardo et al., arXiv:1908.00518

*** D.S. Akerib et al., arXiv:1709.00800

Applications of Xenon-Doped Argon

- WIMP dark matter detection
 - Darkside-20K / GADMC
 - Especially important for extending the reach of ionization-only analysis
- Neutrino physics via the CEvNS channel^{*}
 - Sterile neutrino searches
 - Neutrino magnetic moment searches
 - Non-standard interactions and new light mediators
 - Flavor-blind observation of supernovae, including potential insight into the neutrino mass hierarchy^{**}
- Anti-proliferation technology
 - Reactor fuel cycle monitoring with CEvNS^{***}

Low energy nuclear recoils

Energy spectra are weighted toward lower energies.

Small ionization signal improvements result in large sensitivity gains.

- Large-Scale argon TPC improvements^{****}
 - Shift liquid scintillation light to more easily sensed wavelength
 - Narrower timing of liquid scintillation light
 - Reduced Rayleigh scattering of scintillation light
 - Increased charge yield?

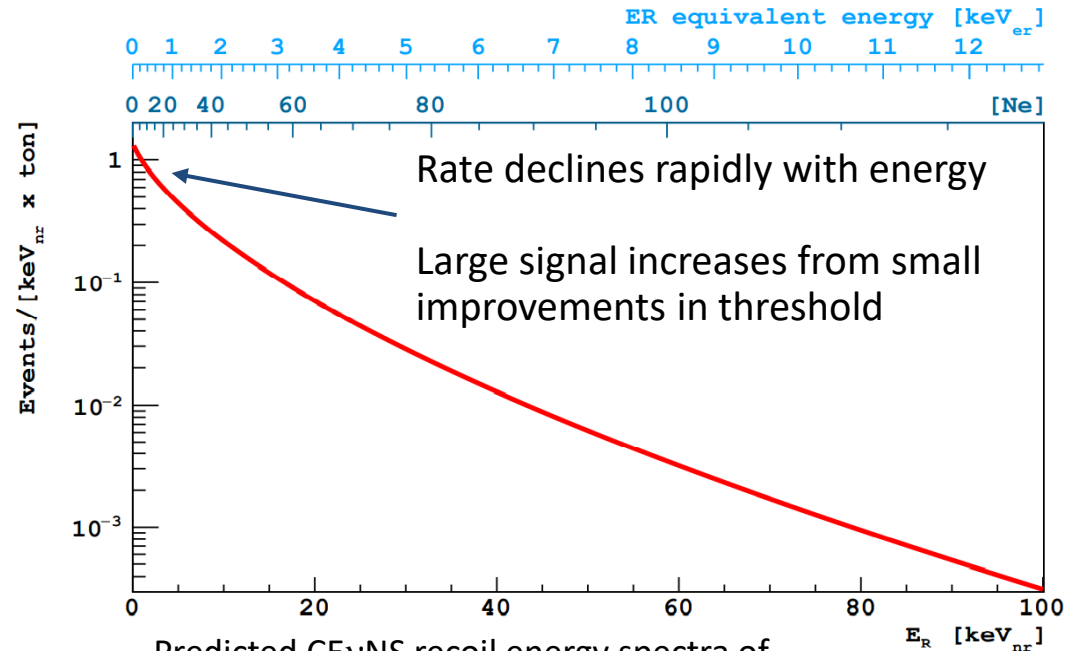
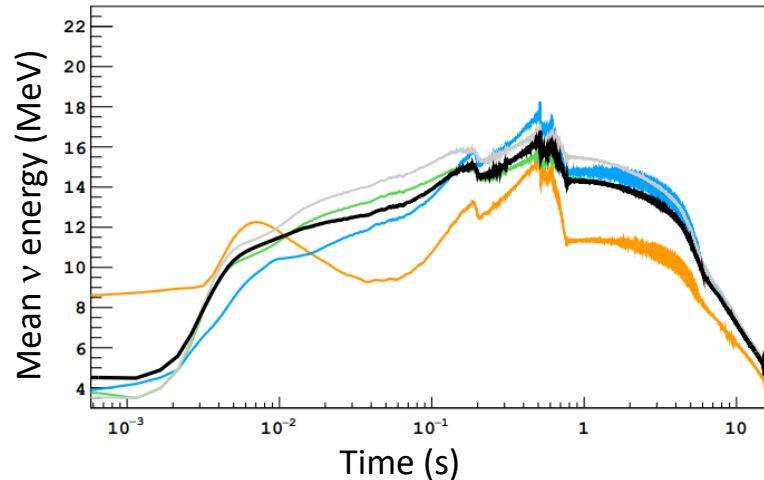
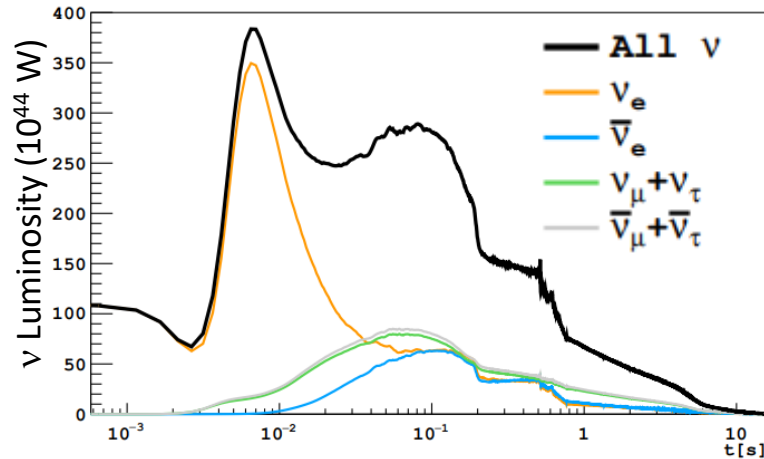
High energy hadrons and leptons

Simplify scintillation optical signal channel

^{*} O.G. Miranda et al., arXiv:2003.12050 ; L.J. Flores et al. arXiv:2002.12342 ; C. Blanco et al. arXiv:1901.08094

^{**} P. Agnes et al., arXiv:2011.07819 ; ^{***} C. Hagmann and A. Bernstein, arXiv:nucl-ex/0411004 ; ^{****} D. Whittington, JINST 11 C05019 (2016)

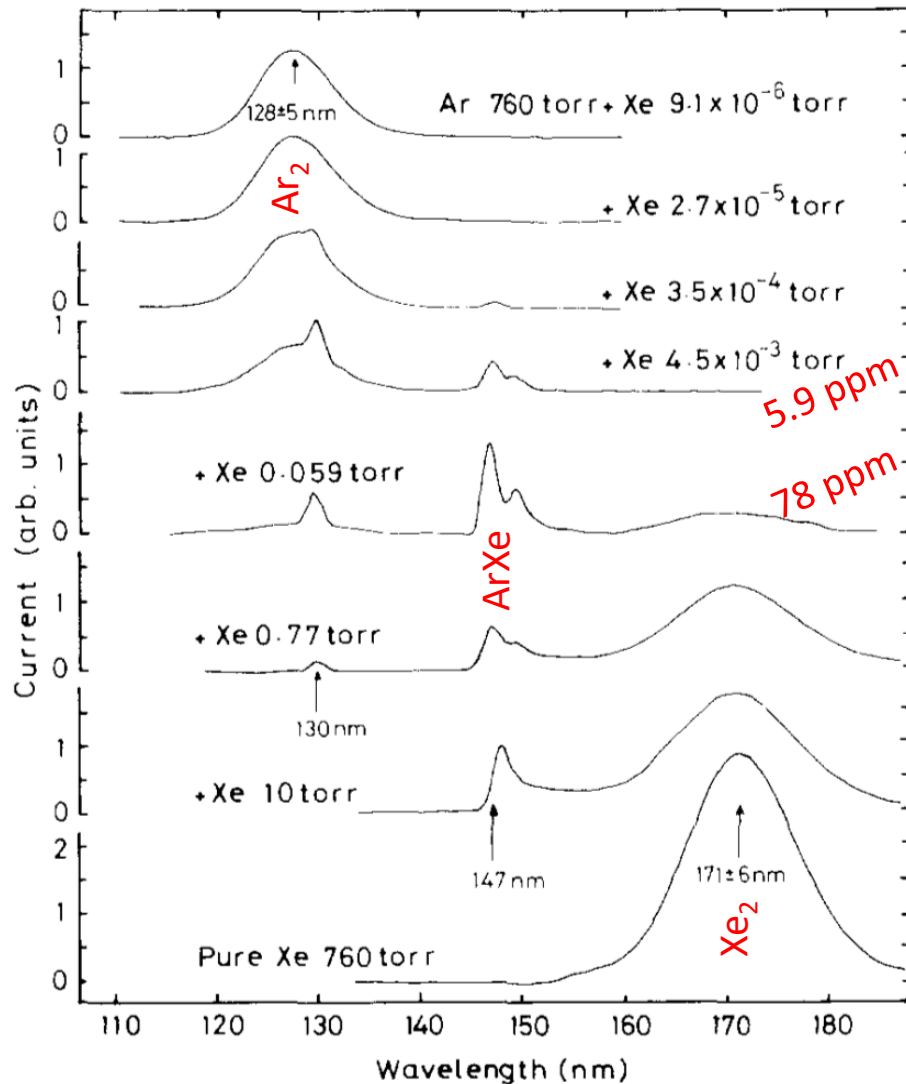
Motivation – Core-Collapse Supernova Detection through CEvNS



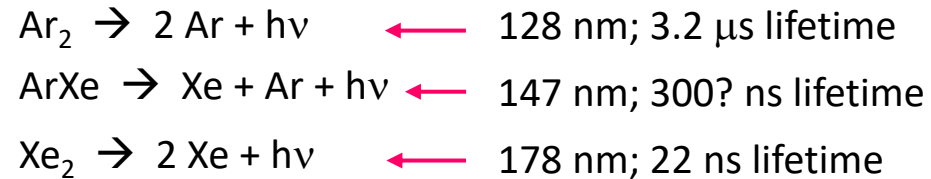
Predicted CEvNS recoil energy spectra of LAr from a core-collapse supernova of 27 solar masses at 10 kiloparsecs*

*DarkSide-20K Collaboration JCAP **03**, 04350 (2021) arXiv:2011.07819

Energy Transfer in Ar Xe Gas Mixtures



Emission spectra of xenon-doped argon gas mixtures at 1 atm in a gas proportional counter*



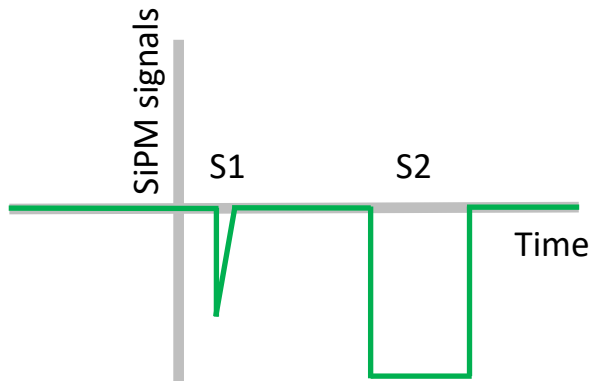
We expect most of the S2 light will be wavelength shifted to 147 nm by ~50 ppm of Xe addition to Ar gas.

QE of 147 nm detection is about 60% higher than 128 nm detection with new VUV Hamamatsu SiPMs

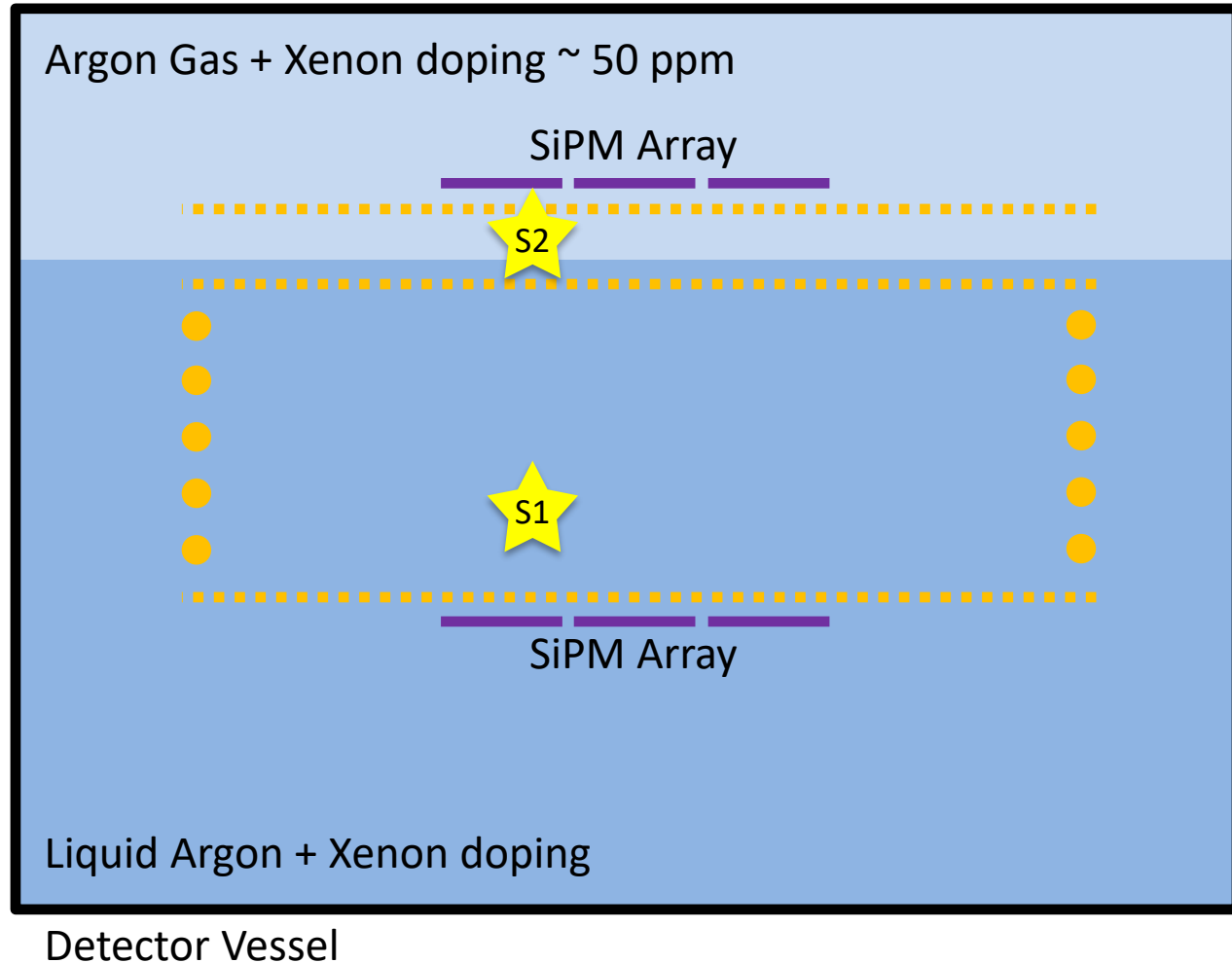
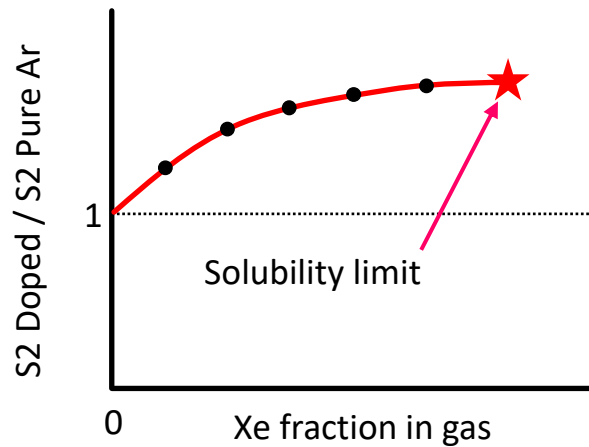
* T. Takahashi et al.
NIM **205** 591-596 (1983)

Yuto Ohashi, Hamamatsu Photonics K.K.
CHEF Conference (2019)

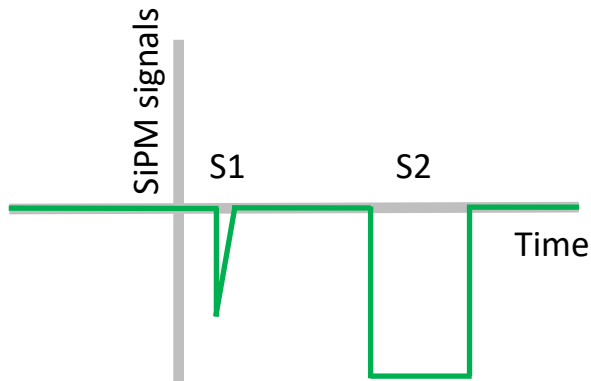
Xenon-Doped Argon S2 Experiment



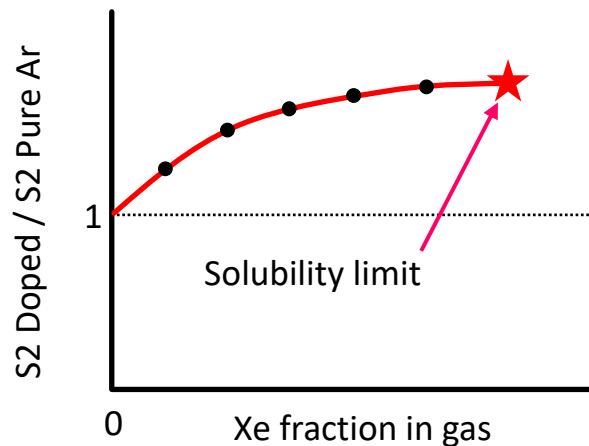
Anticipated data



Xenon-Doped Argon S2 Experiment



Anticipated data



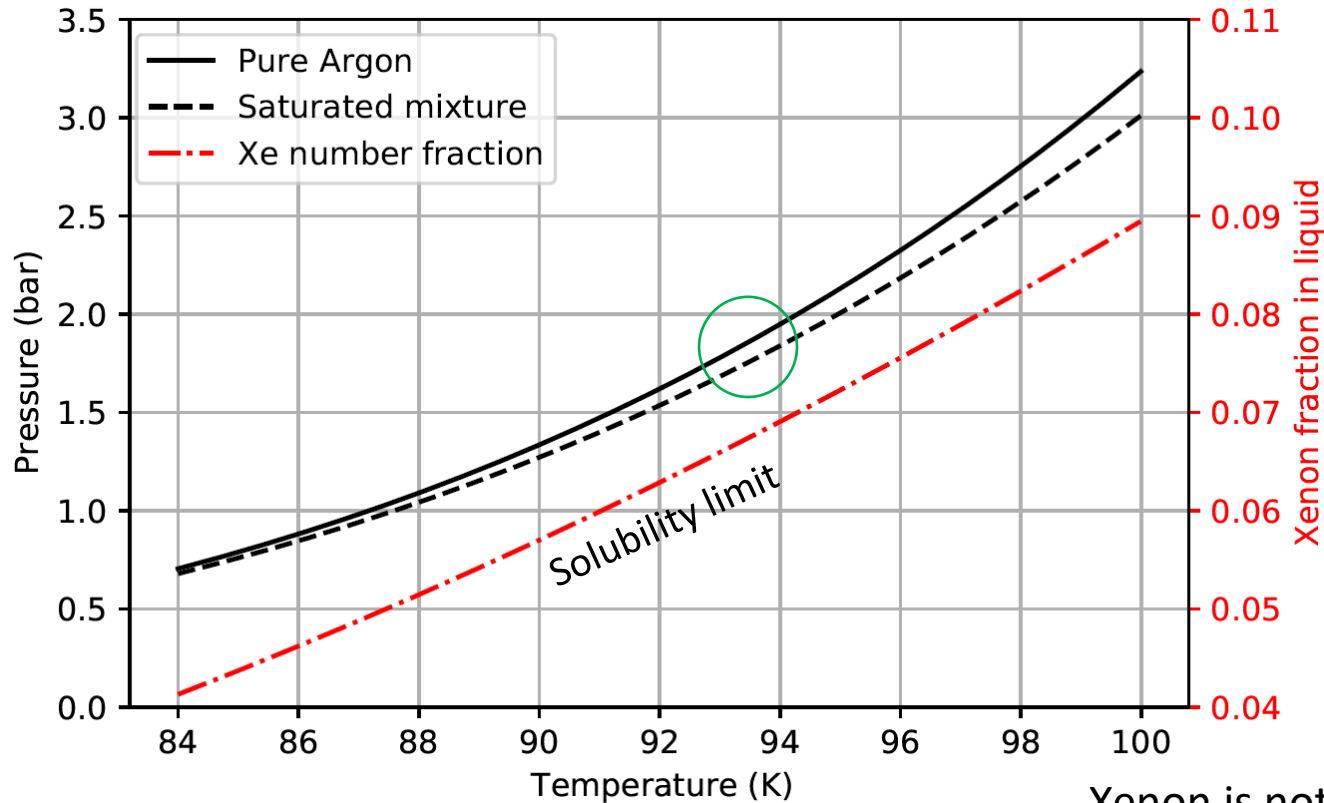
Argon Gas + Xenon doping ~ 50 ppm

How do we provide this environment?

Liquid Argon + Xenon doping

Detector Vessel

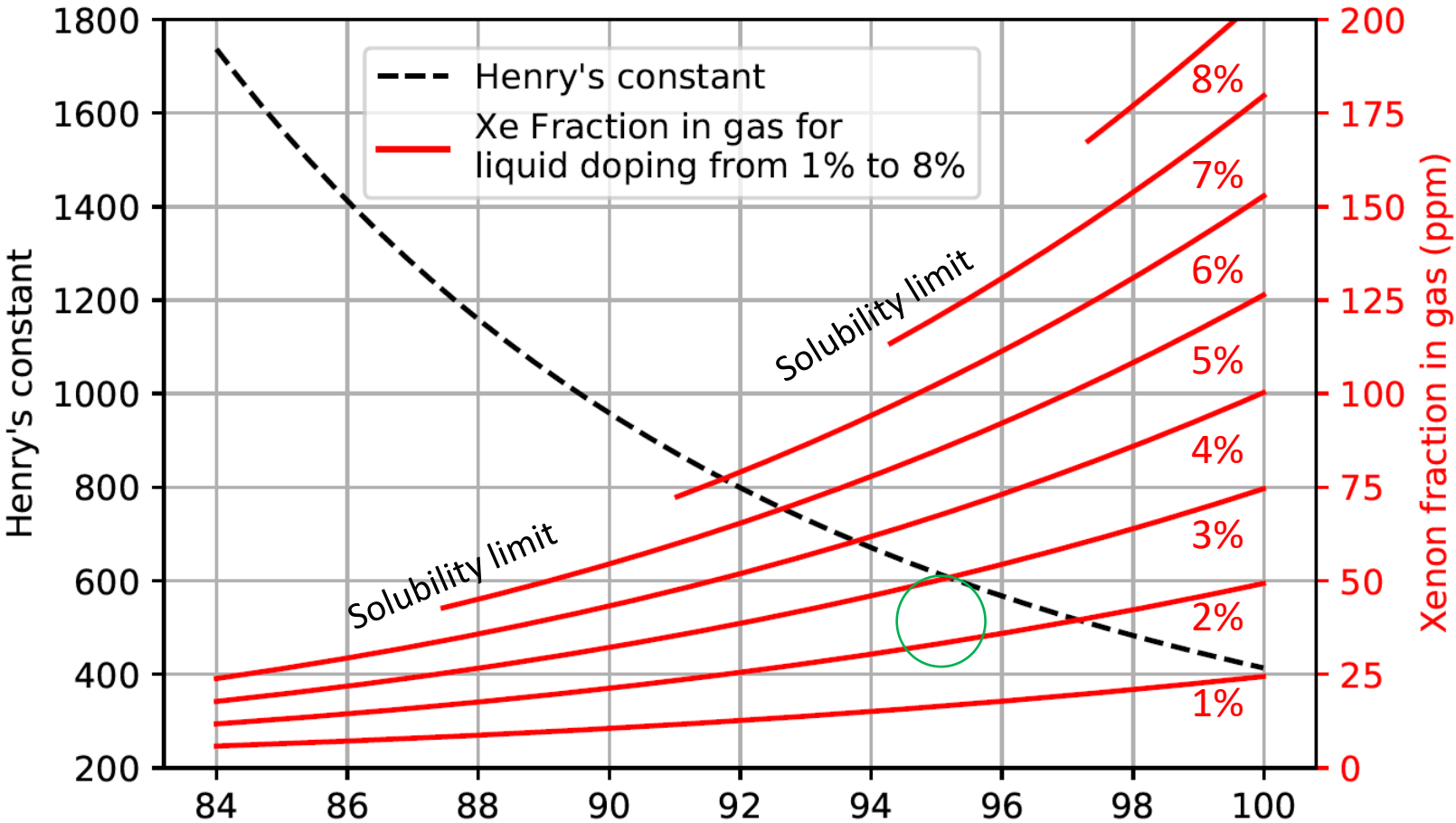
Xenon-Doped Argon Thermodynamics



Xenon is not miscible with argon at low temperatures

Unwanted solid formation may occur

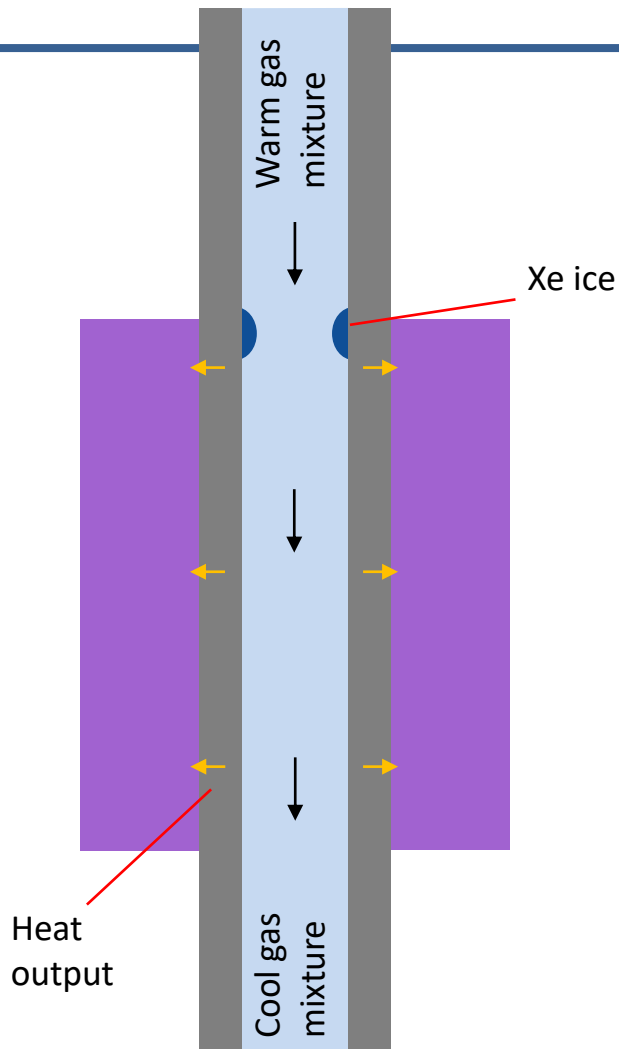
Xenon-Doped Argon Thermodynamics



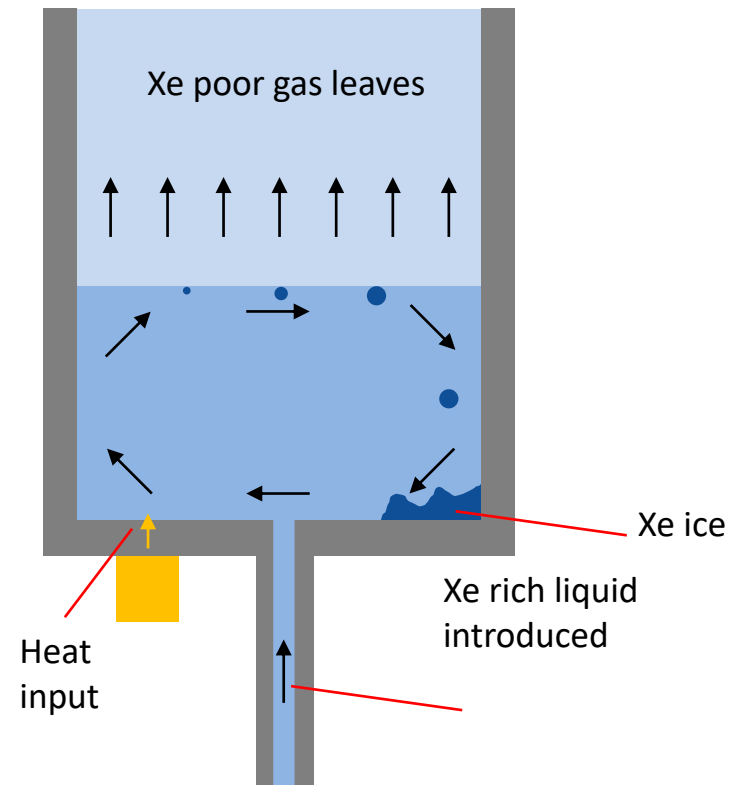
Xenon strongly partitions into the gas

Unwanted distillation may occur

Mixture Instability Mechanisms

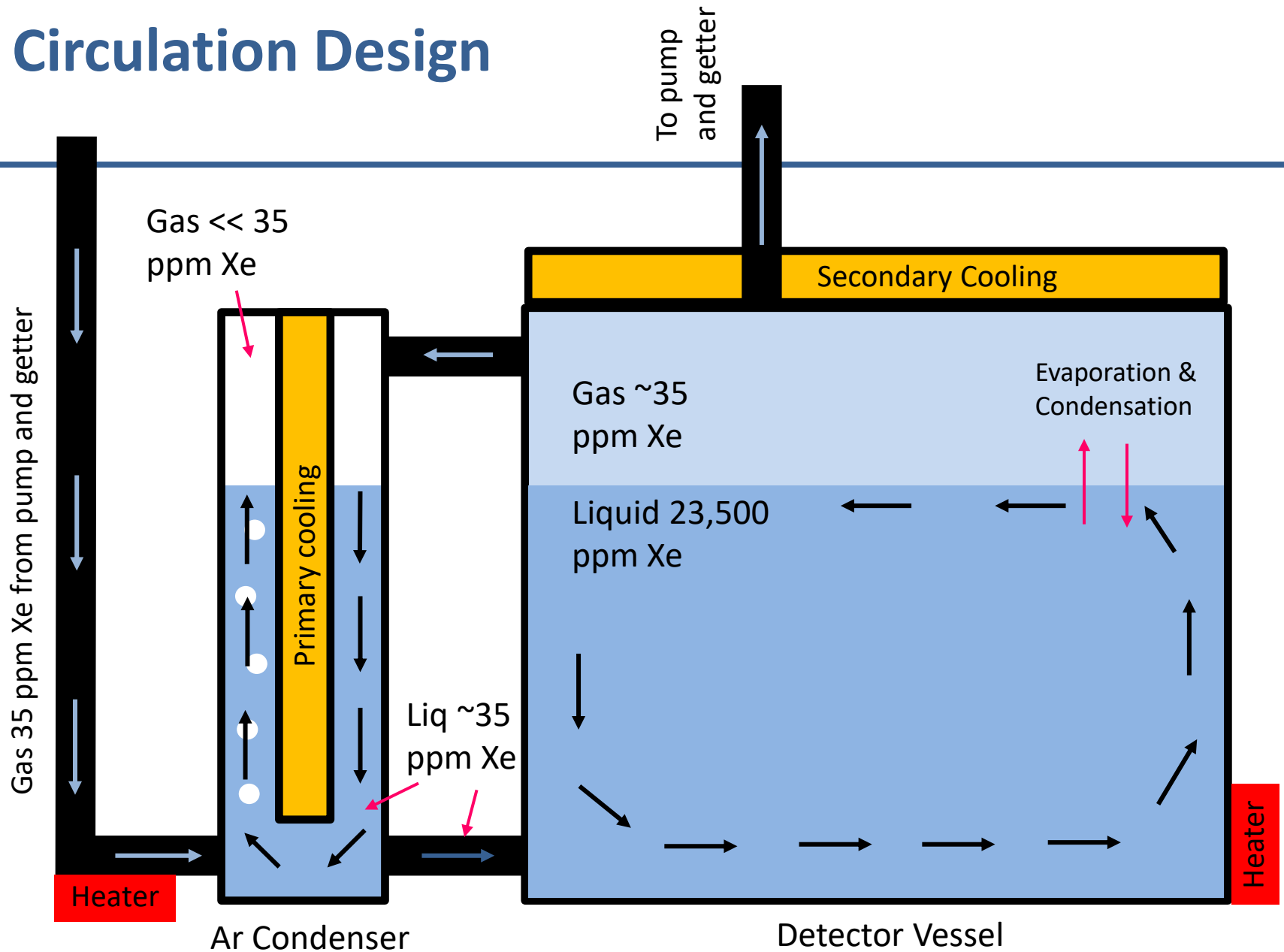


Gas cooling instability

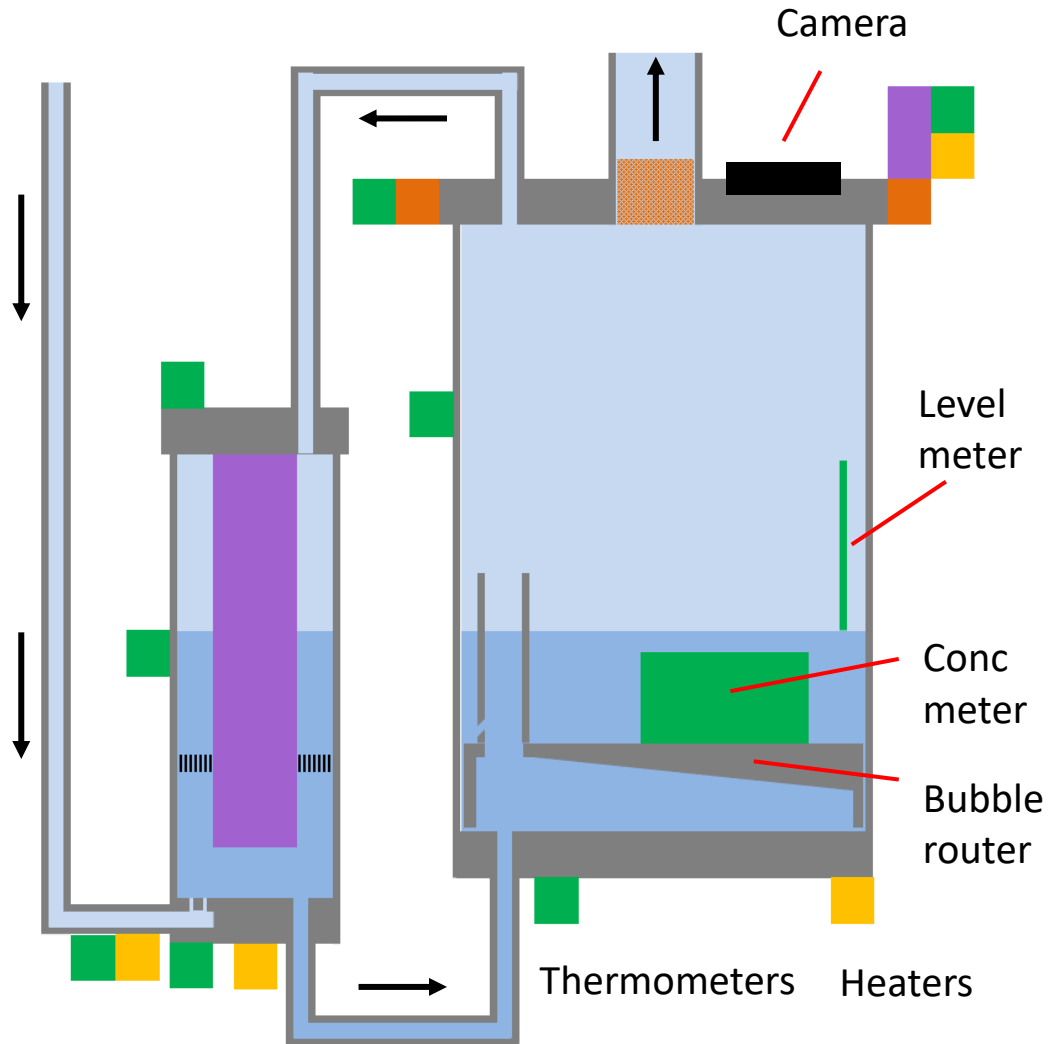


Distillation instability

Circulation Design

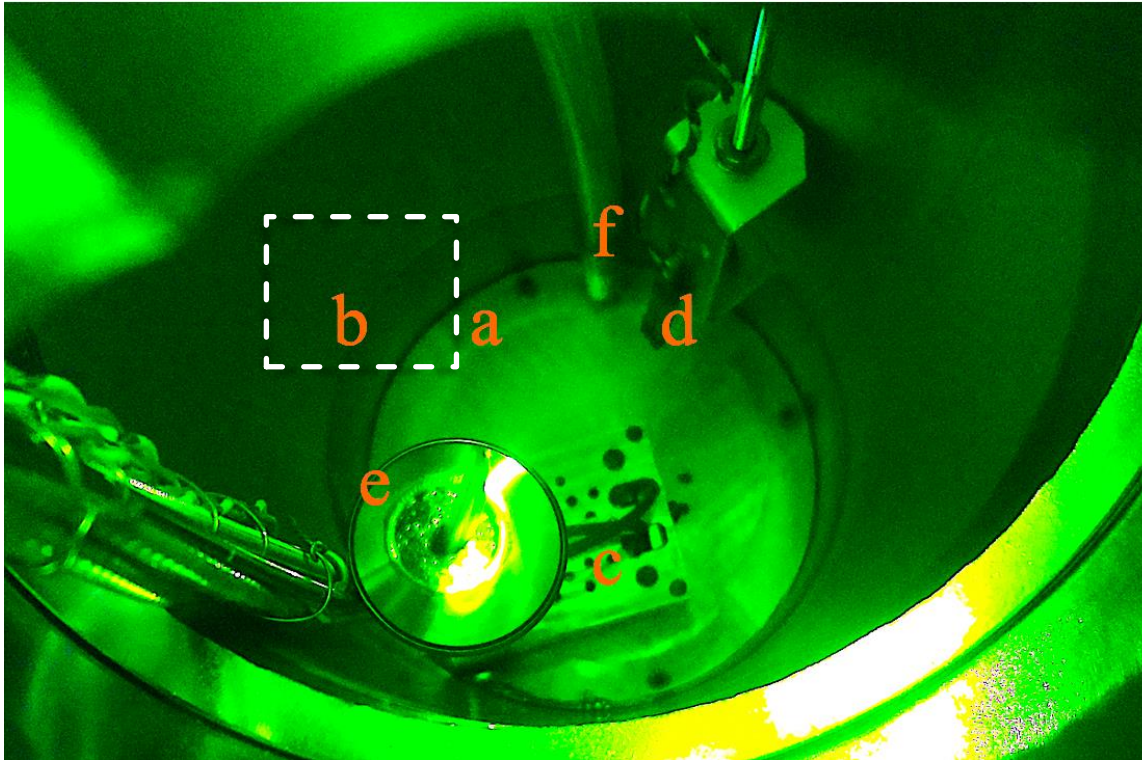


Circulation Design



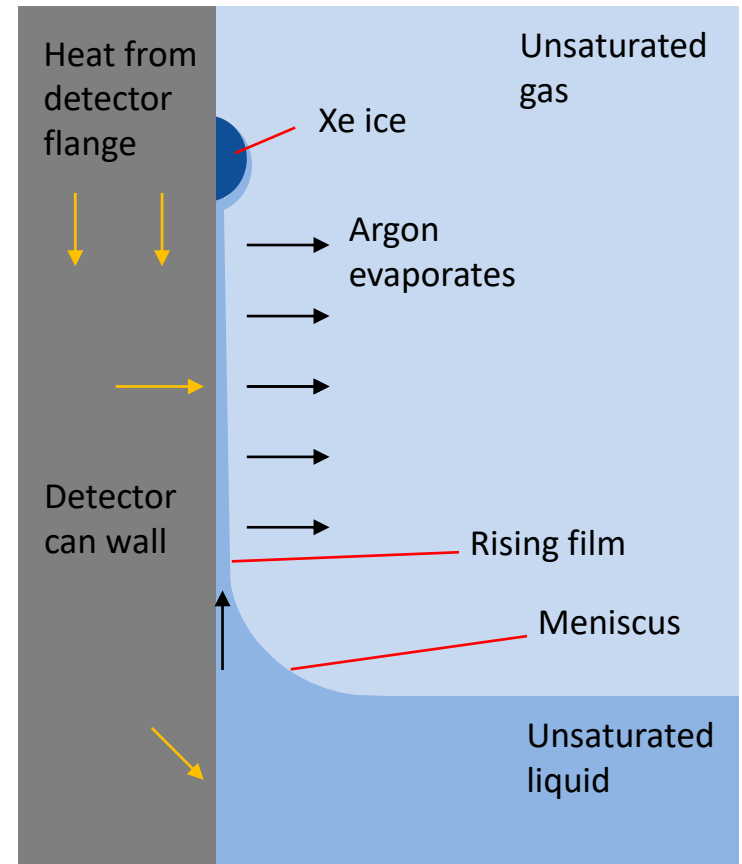
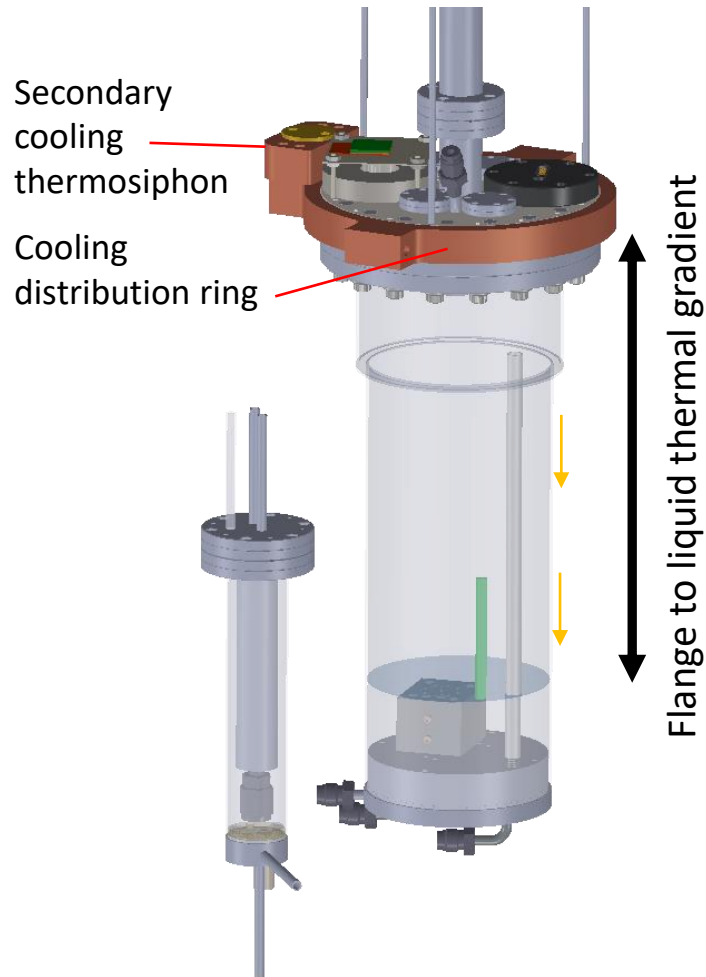
Camera View of Detector Inside

Looking down into detector can



- a) Bubble routing plate
- b) Liquid level
- c) Capacitive concentration meter
- d) Level meter
- f) Bubble routing tube
- e) Mirror showing cable entry

Wicking Separation Mechanism



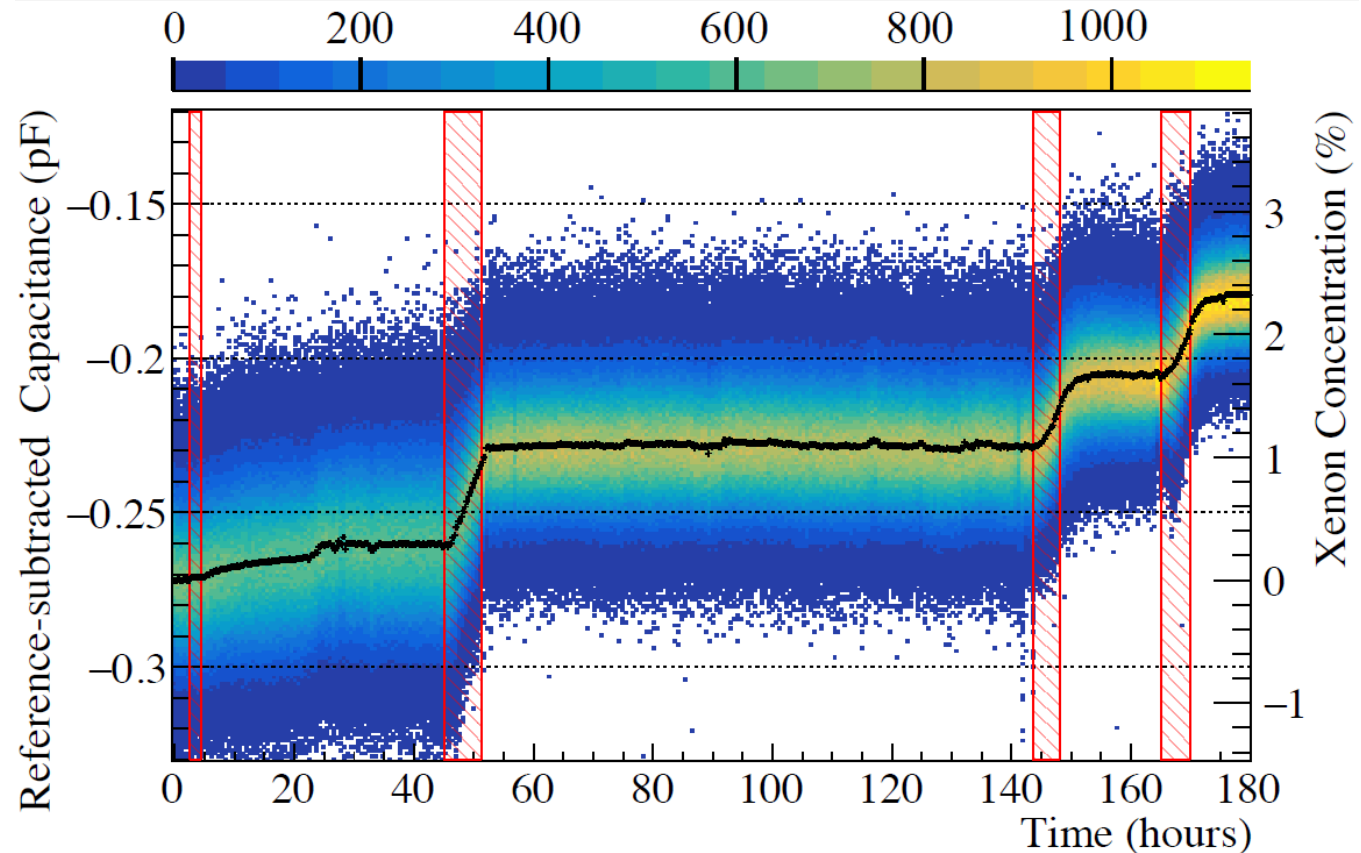
Hypothesized Wicking Separation Mechanism

Doping Xenon Gas Into Liquid Argon

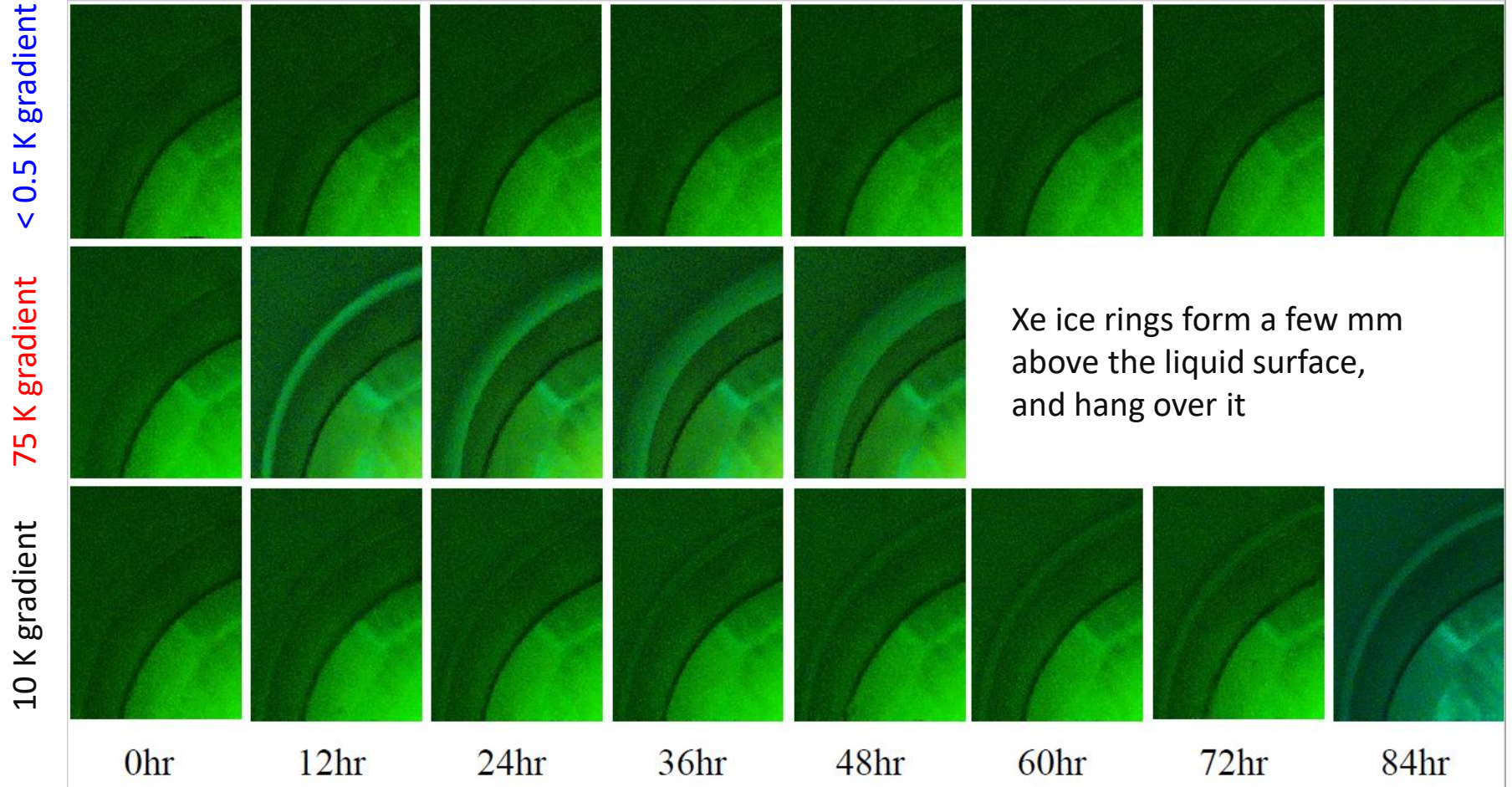
Four doping periods, each with 0.6% Xe gas stream introduced into the condenser at 1.5 SLPM

Xe appears promptly in the main bath when the detector flange temperature is properly adjusted

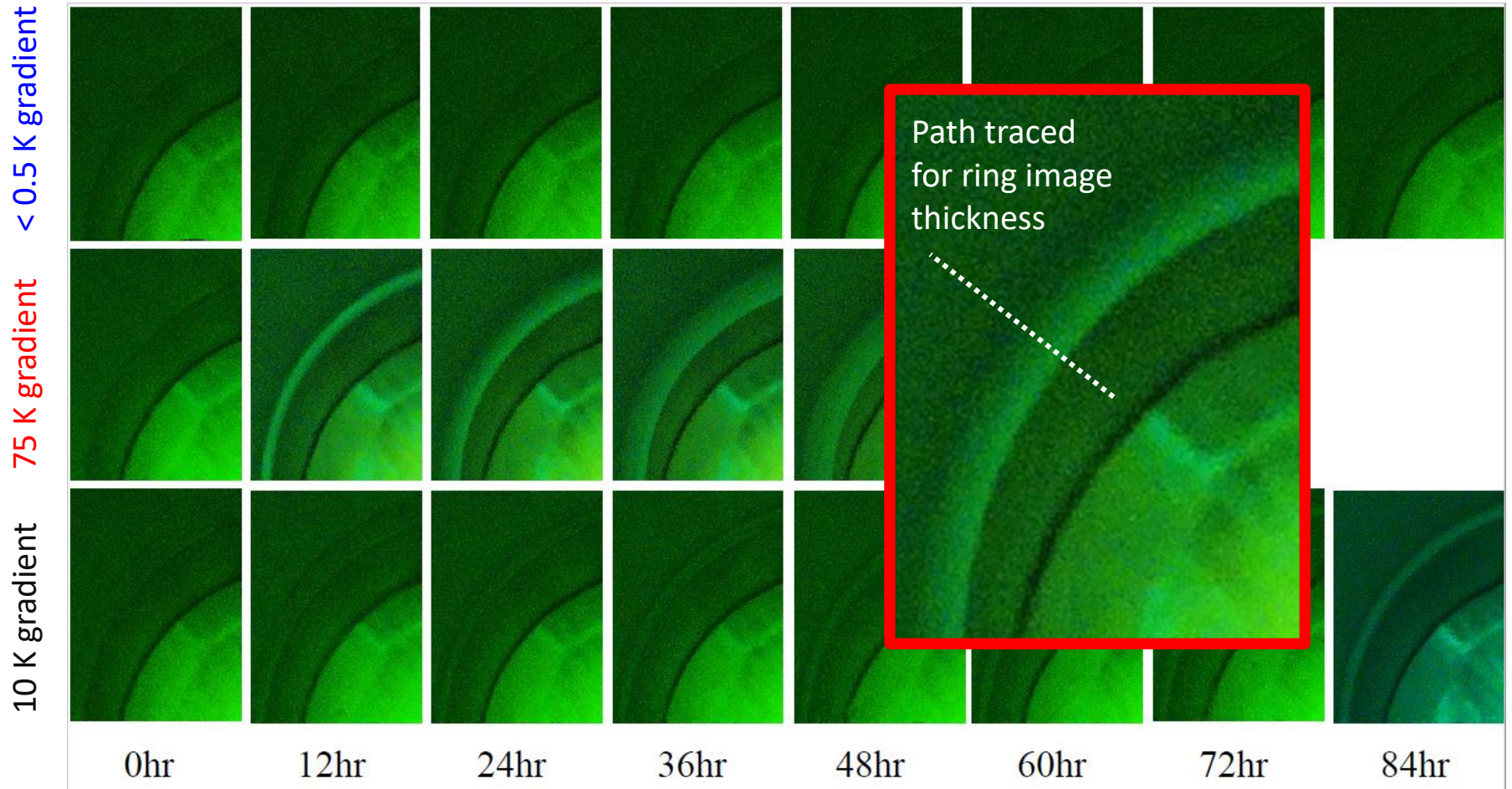
Outgoing gas samples after last doping step show 30 – 50 ppm Xe as measured by RGA



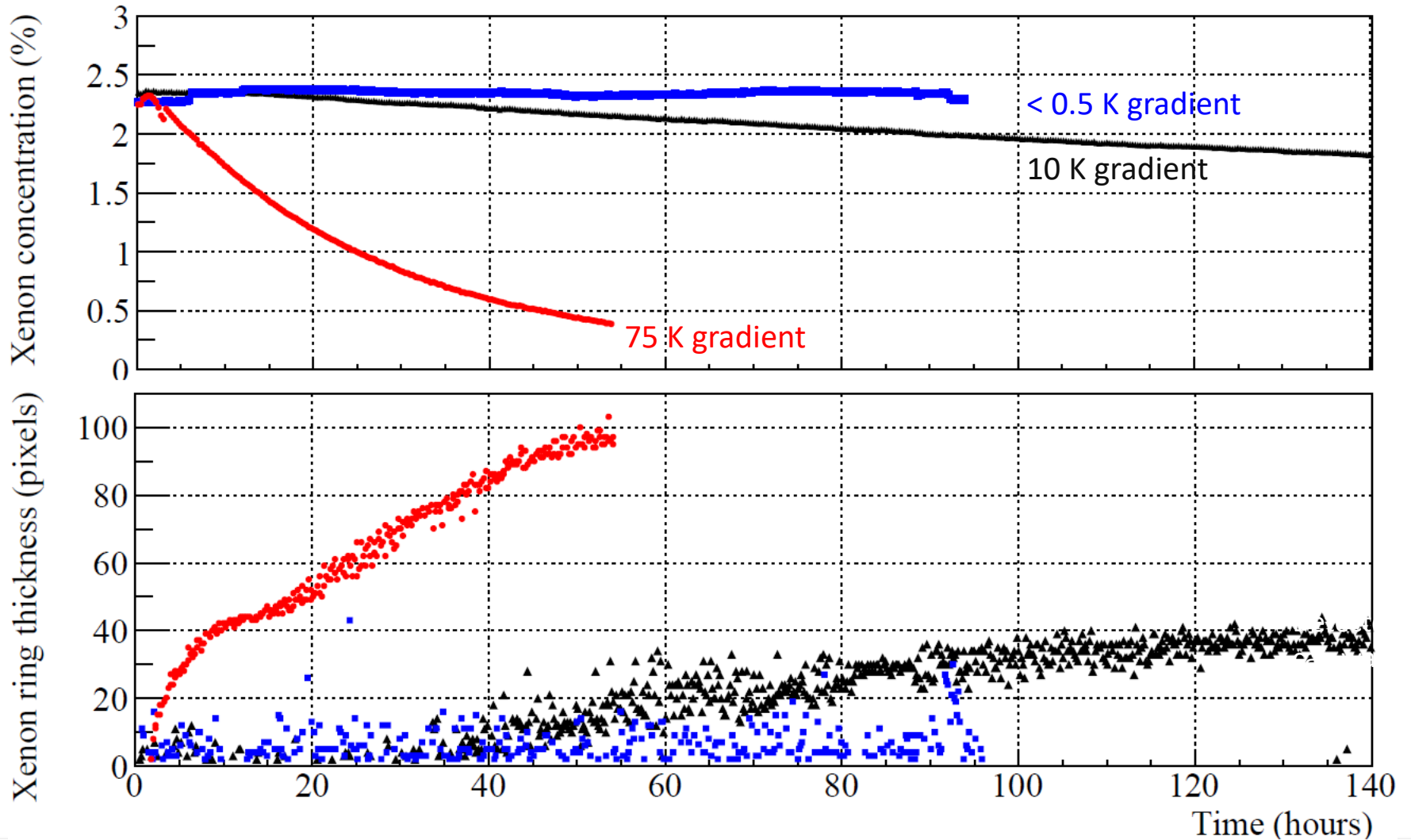
Wicking Separation Under Different Gradients



Wicking Separation Under Different Gradients



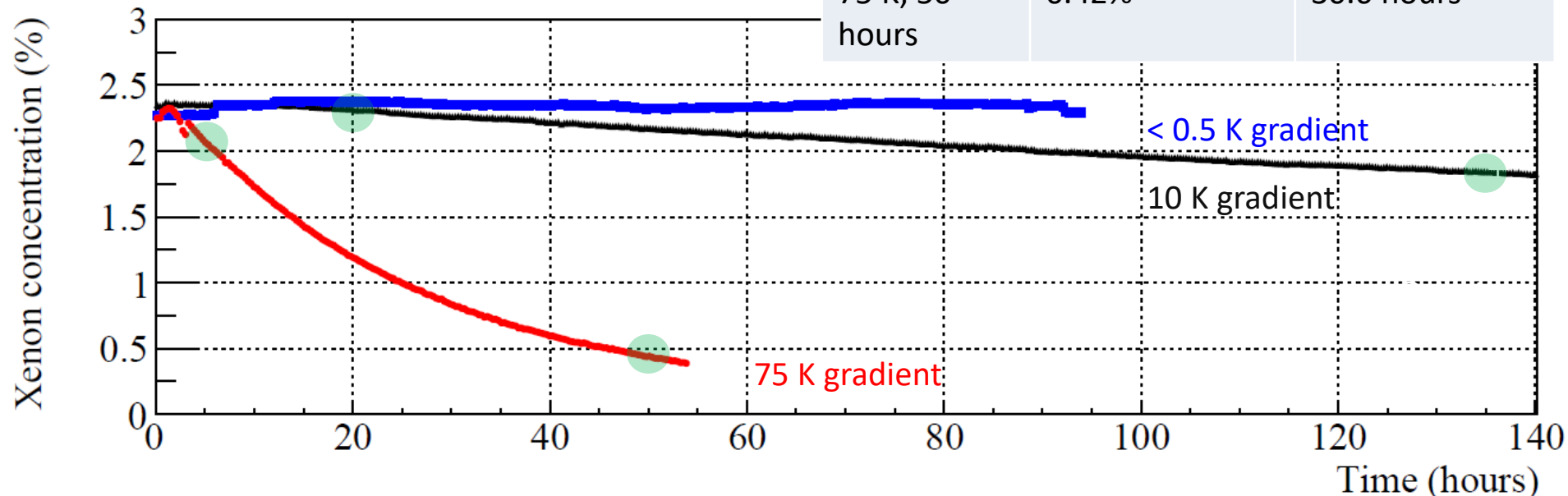
Wicking Separation



Wicking Separation

Slopes and concentrations verify simple exponential behavior

Condition ΔT , time	Concentration	Timescale, from slope and value of concentration
10 K, 20 hours	2.3%	470 hours
10 K, 135 hours	1.7%	493 hours
75 K, 5 hours	2.08%	27.7 hours
75 K, 50 hours	0.42%	30.6 hours



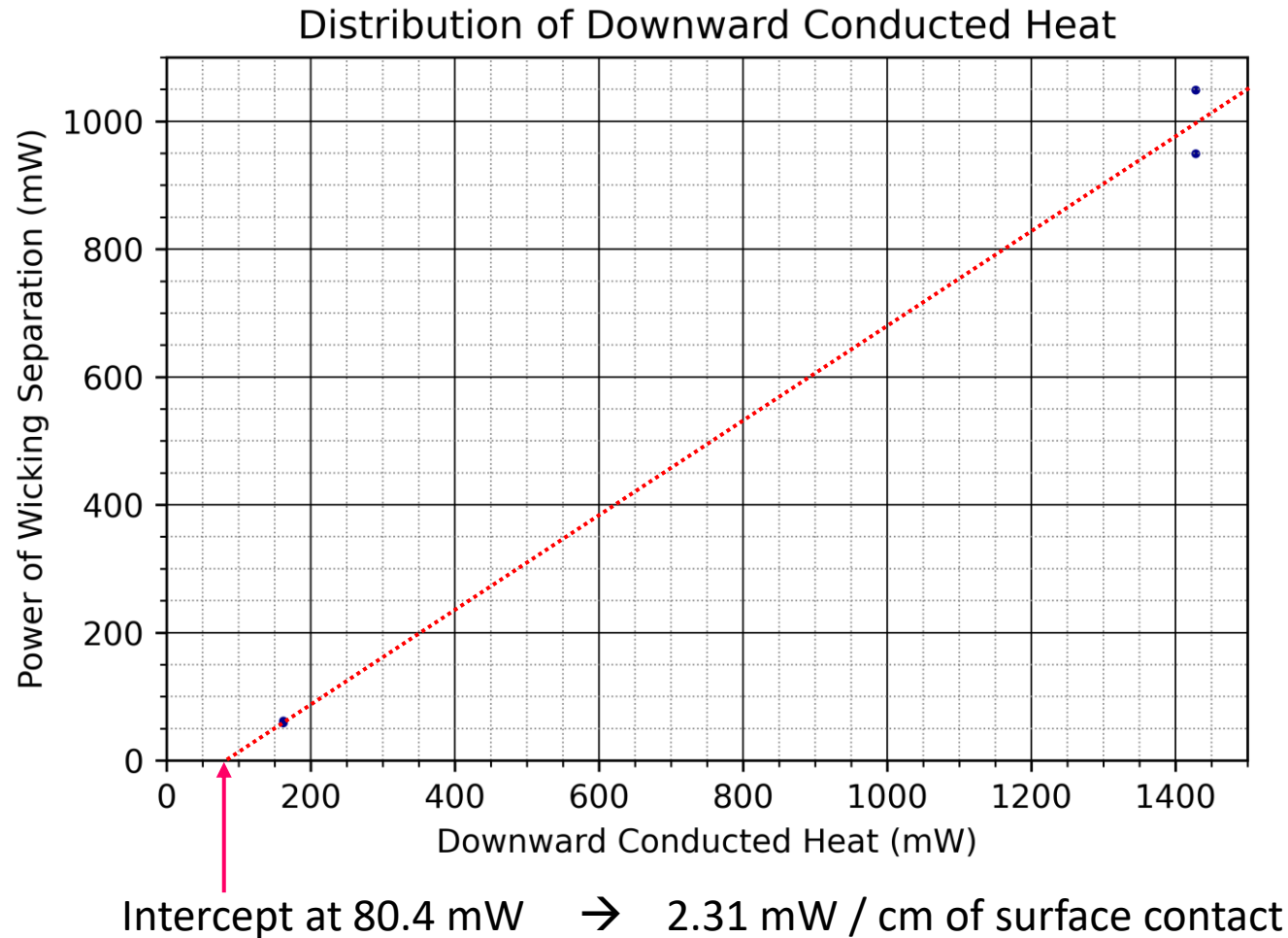
Wicking Separation

Determine from
timescale, liquid
mass, heat of
vaporization

Determine
from ΔT ,
material,
geometry

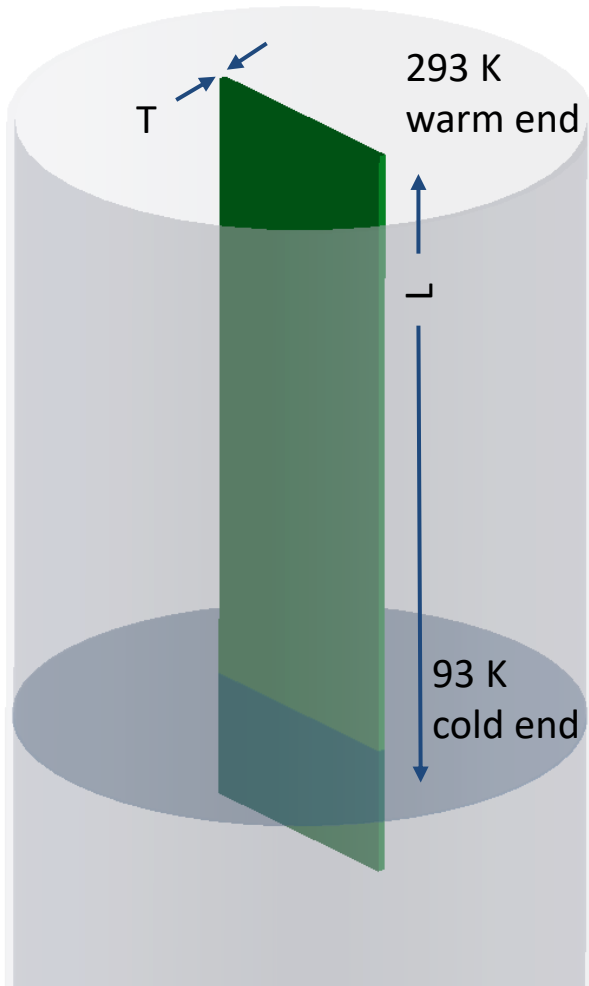
Condition ΔT , time	Concentration	Timescale, from slope and value of concentration	Wicking separation power	Downward conducted heat
10 K, 20 hours	2.3%	470 hours	61.7 mW	162 mW
10 K, 135 hours	1.7%	493 hours	58.8 mW	162 mW
75 K, 5 hours	2.08%	27.7 hours	1049 mW	1428 mW
75 K, 50 hours	0.42%	30.6 hours	949 mW	1428 mW

Downward Conducted Heat and Wicking Separation



Thin Element Descending From Room Temperature

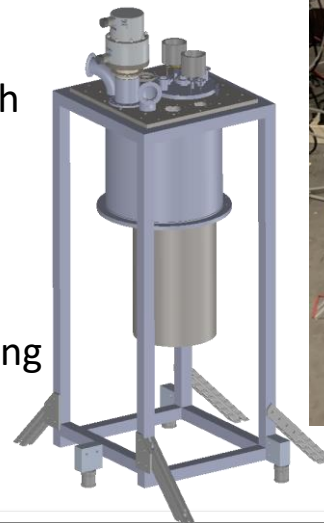
Minimum aspect ratios of vertical thin elements thermally anchored to room temperature and submerged in the doped liquid mixture. *This is approximate and speculative, and only to guide further testing.*



Material	Thermal conductivity at 200 K $W / (m K)$	Minimum L / T
304 SS	12.63	5460
G10 (warp direction)	0.6741	292
PTFE	0.2672	116
Kapton	0.1749	76

Conclusion

- Predictable, stable xenon doping of argon liquid and gas:
 - Circulation design allows rapid xenon introduction and mixing.
 - We can establish stable concentrations of xenon up to 2.35 % in liquid
 - Gas sampled from above the liquid contains tens of ppm Xe as measured by RGA, in agreement with expectations from Henry's law.
 - Unwanted solid xenon formation can be controlled with proper thermal design.
- Coming up:
 - Measurement of improvement in S2 light from Xe doping
 - Measure changes to ionization yields



The CHILLAX detector



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Prepared by LLNL under Contract DE-AC52-07NA2

Backups



Enabling Technology – ^{39}Ar depleted argon

Depleted argon infrastructure

- Urania plant (330 kg / day) under construction at the Kinder-Morgan Doe Canyon facility, Colorado, USA*

Argon is separated from CO_2 wells and purified

^{39}Ar reduced by a factor of 1400 relative to atmospheric sources.

- ARIA project: Cryogenic distillation column for argon isotope separation. Under construction in the Seruci Mine, Sardinia, Italy**

350-meter cryogenic distillation column

^{39}Ar reduction by a factor of 10 *per pass*



Prototype ARIA distillation column

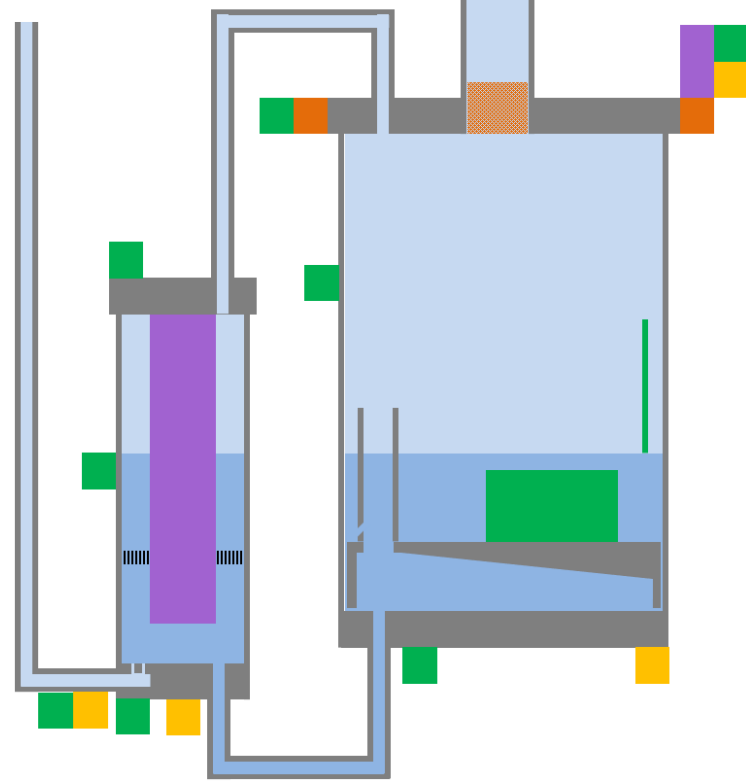
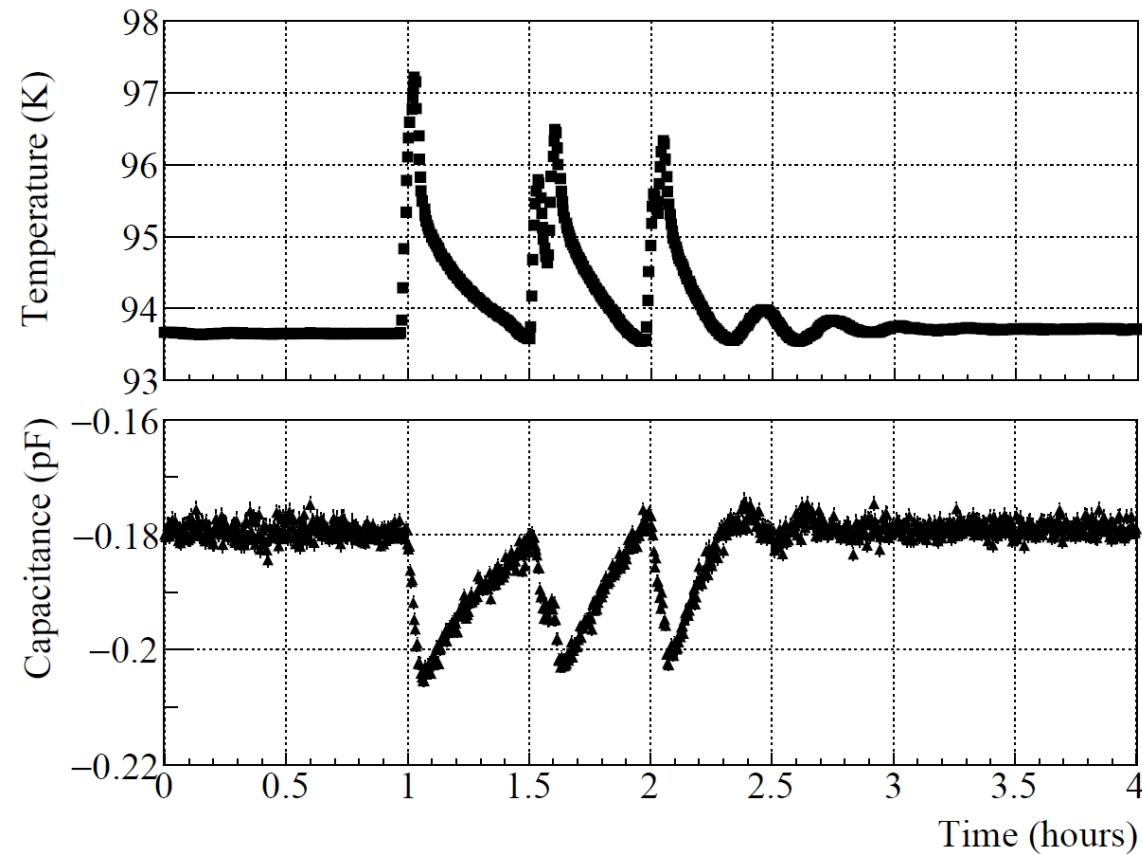
* W. Bonivento doi:10.1088/1742-6596/1468/1/012234

** Agnes, P. *et al.*, Eur. Phys. J. C 81, 359 (2021)

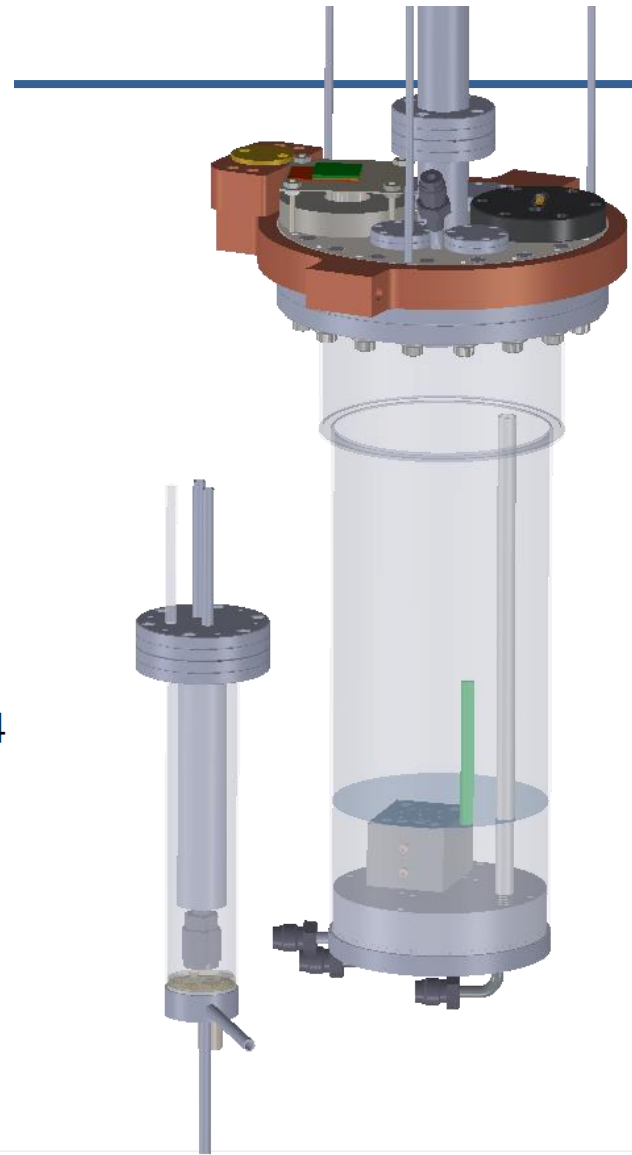
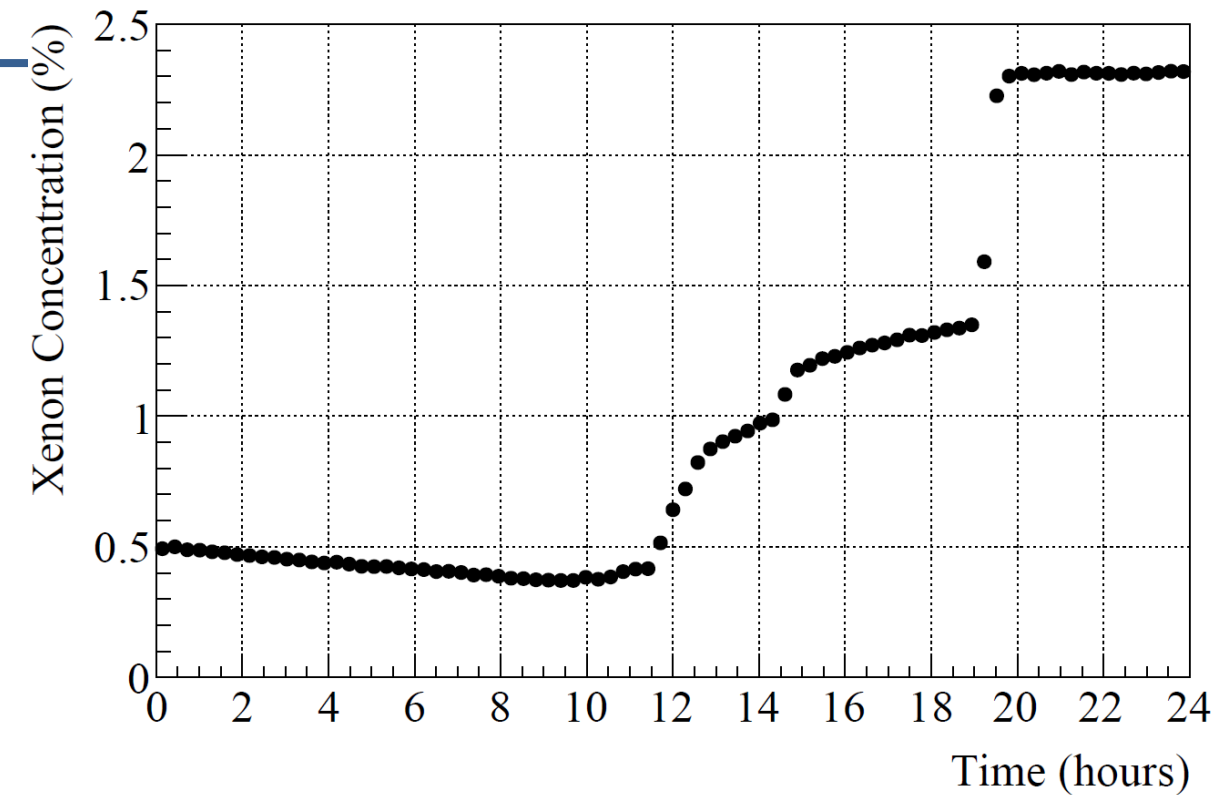
Circulation Design



Mixing Verification Test



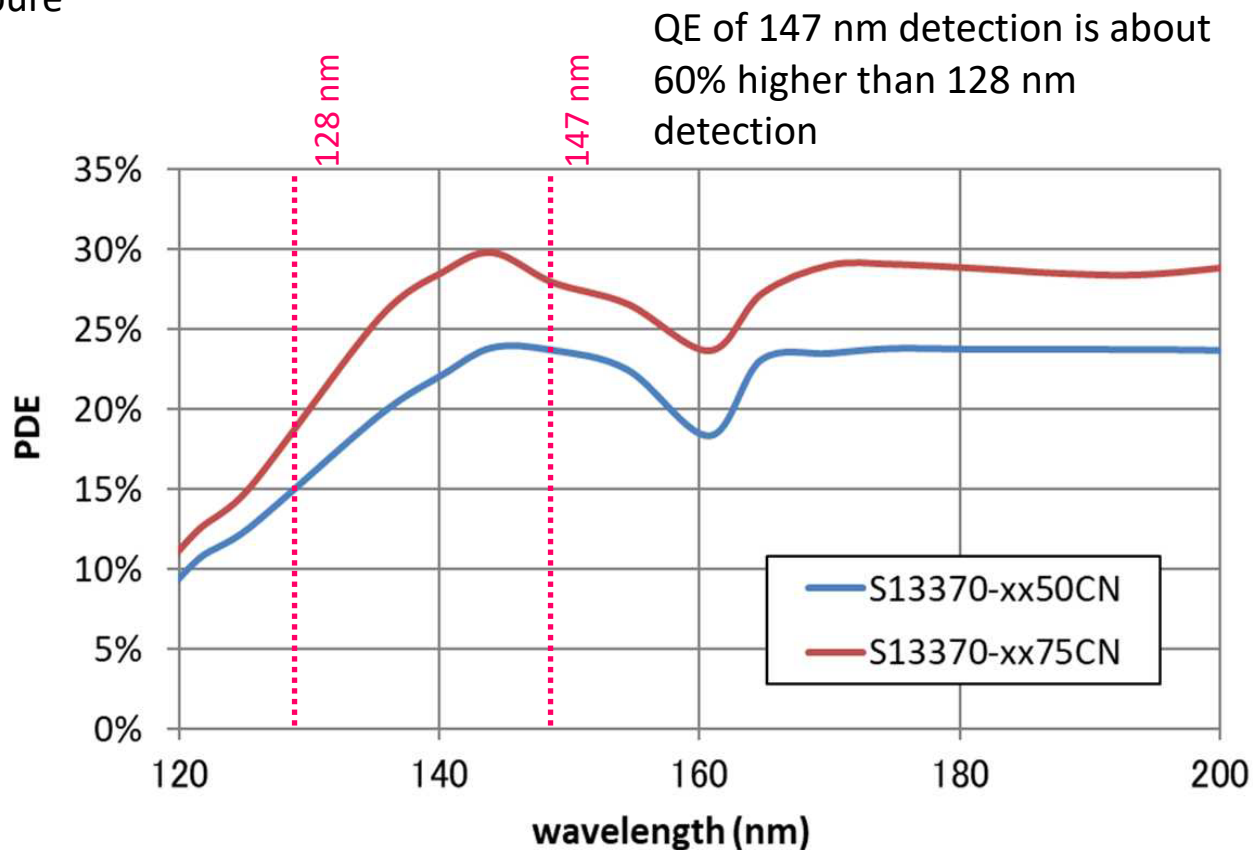
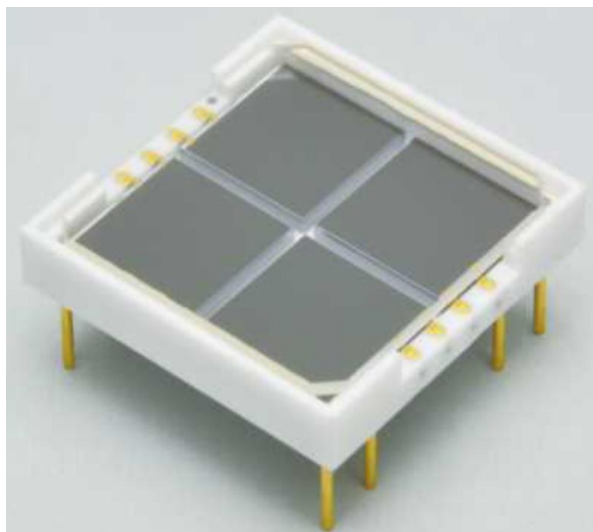
Ice Washdown



Enabling Technology – VUV SiPMs

VUV SiPM development

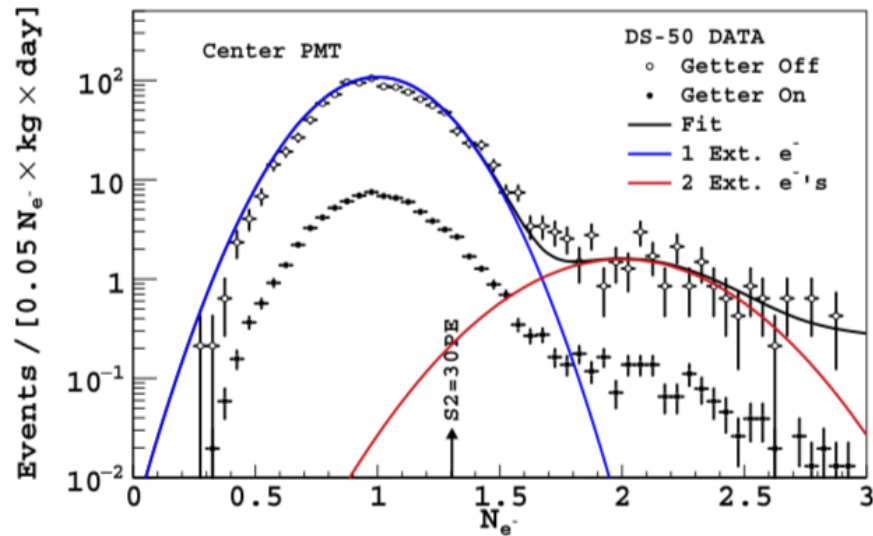
- Durable, compact, and radiopure relative to PMTs
- Numerous cryogenic amplification schemes*



* M. D’Incecco et al. arXiv: 1706.04213 ; A. Falcone et al., arXiv: 2001.09051

Yuto Ohashi, Hamamatsu Photonics K.K. CHEF Conference (2019)

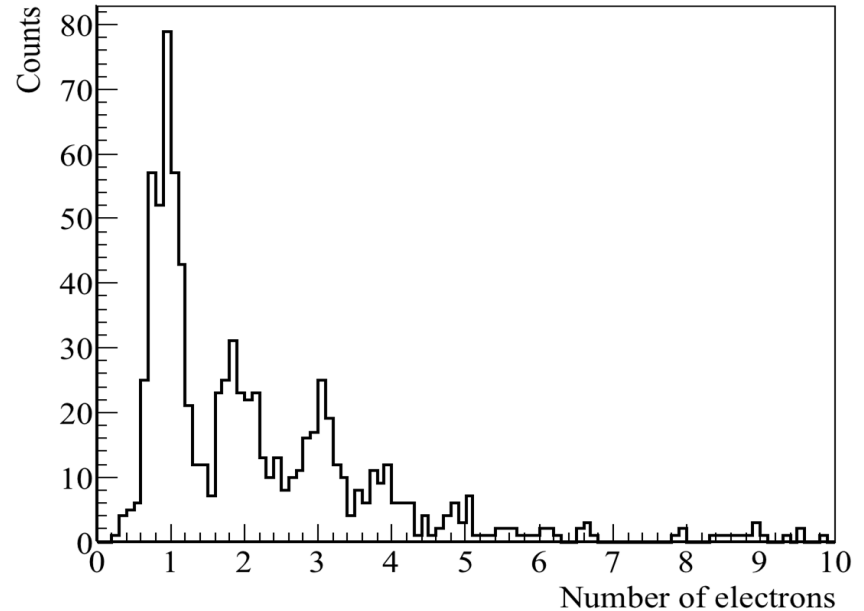
Single electron spectra: Xenon and Argon



DarkSide50 SE spectrum
PRL, 121, 081307 (2018)

23 PMT photoelectrons /
extracted electron

Measurement of wavelength-
shifted argon S2 light



XeNu detector SE spectrum
J. Xu, Magnificent CE νNS workshop (2020)

72 PMT
photoelectrons /
extracted electron

Direct measurement of
xenon S2 light

Detector design



Solubility considerations

Extrapolating to
 $100 / T = 1.054$ from plot at right
 Predicts $n^{\text{Sat}} = 7.1\%$ at 2 bar

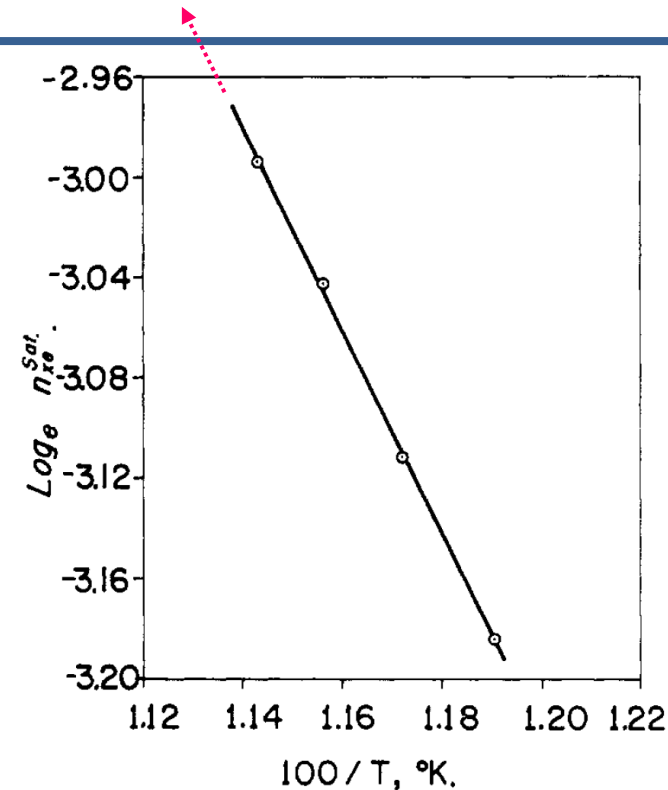
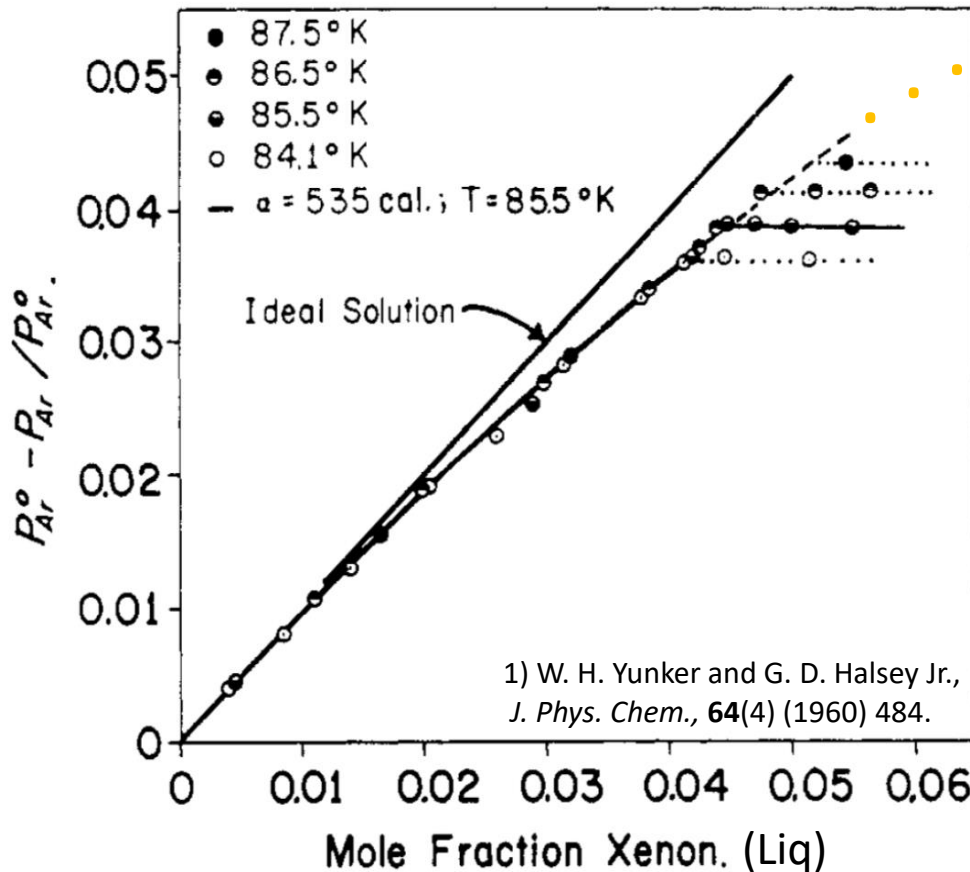
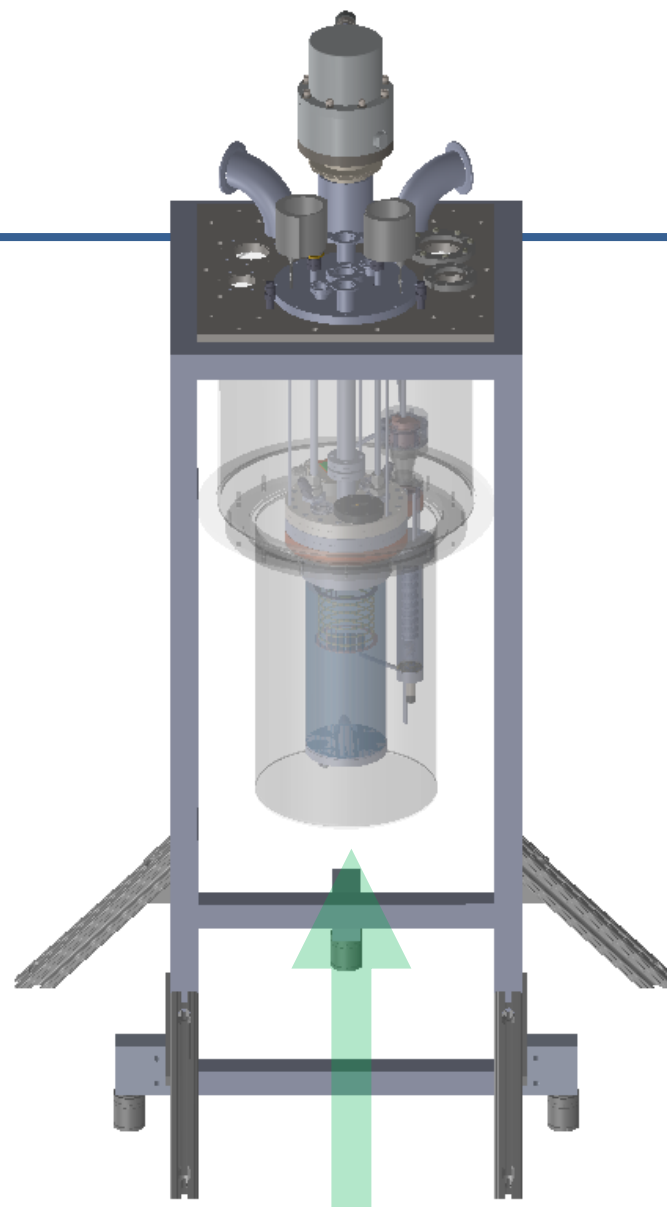
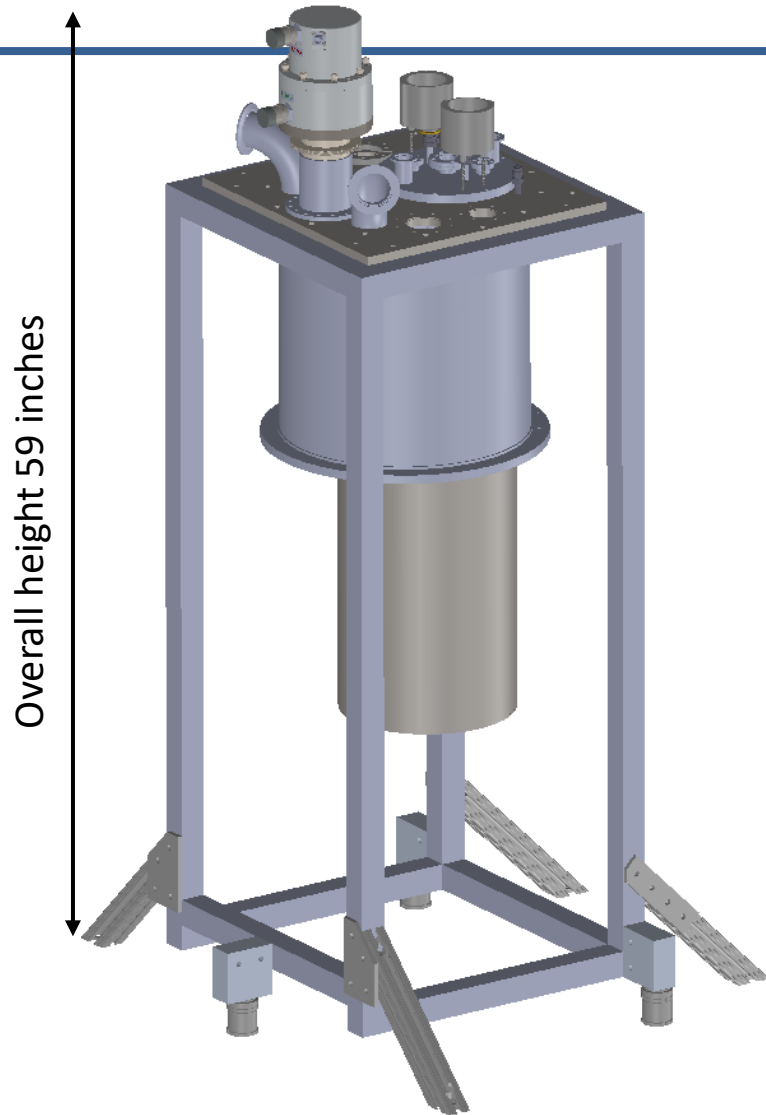


Fig. 2.— $\text{Ln } n_{\text{Xe}}^{\text{Sat}}$ vs. $100/T$ °K.

Temp. (°K.)	α (cal./mole)
84.0	560 ± 55
85.5	536 ± 39
86.5	504 ± 39
87.5	504 ± 20
Av.	535 ± 58

Detector design



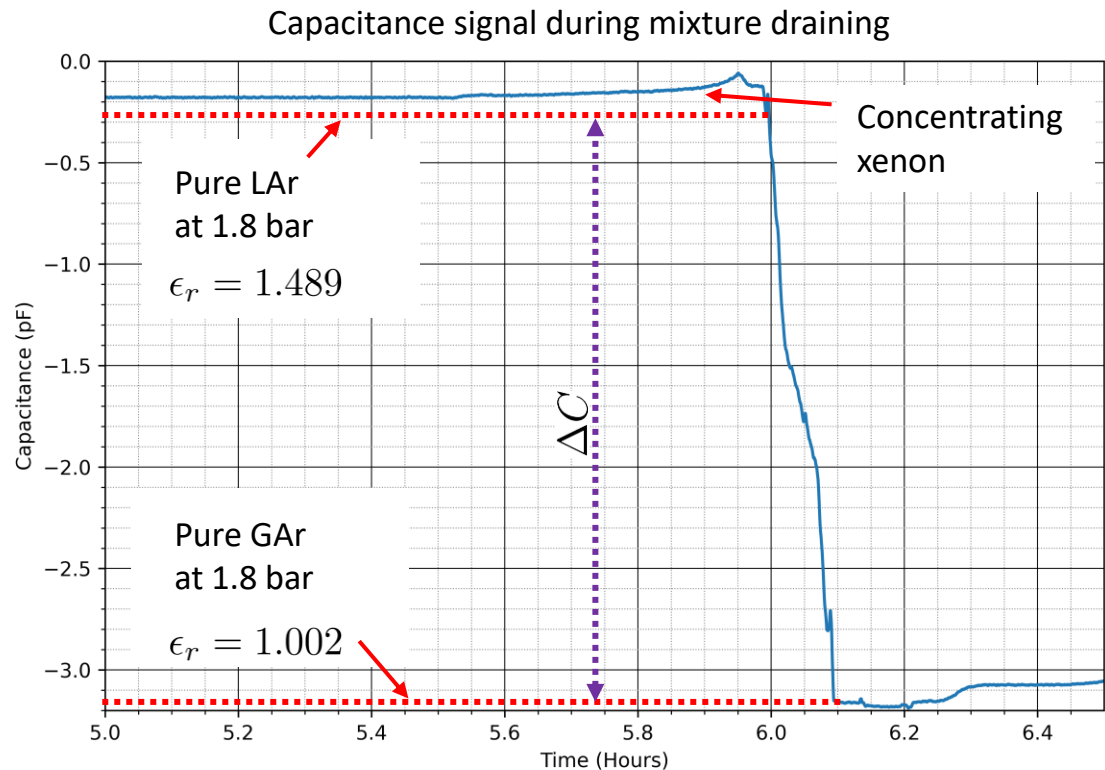
Low obstruction path for neutron scattering measurements

Capacitive Sensitivity to Temperature Fluctuations

Values of ϵ_r for non-standard densities can be produced by the C-M equation and the literature value of 1.505 for 1 bar on the vapor curve

$$C = \epsilon_r \cdot C_{\text{vac}}$$

$$C_{\text{vac}} = \Delta C / \Delta \epsilon_r = 5.926 \text{ pF}$$



Capacitive Sensitivity to Temperature Fluctuations

Thermal changes in the measured capacitance have two sources:

$$C = \epsilon_r \cdot C_{\text{vac}}$$

$$\frac{dC}{dT} = \frac{d\epsilon_r}{dT} \cdot C_{\text{vac}} + \frac{dC_{\text{vac}}}{dT} \cdot \epsilon_r$$



From liquid
density
changes



From
unwanted
mechanical
changes

Suppose no unwanted sources:

$$\frac{dC}{dT} = \frac{d\epsilon_r}{dT} \cdot C_{\text{vac}} + \cancel{\frac{dC_{\text{vac}}}{dT} \cdot \epsilon_r}$$

$$\frac{dC}{dT} = C_{\text{vac}} \cdot \frac{d\epsilon_r}{dn} \cdot n \cdot n^{-1} \frac{dn}{dT}$$



From draining
measurement



From C-M eq. at 1.8
bar LAr density



Fractional density change of LAr
with temperature at 1.8 bar

$$\frac{dC}{dT} = -16.13 \text{ fF/K}$$

Prediction

Capacitive Sensitivity to Temperature Fluctuations

Prediction from
draining and density :

$$\frac{dC}{dT} = -16.13 \text{ fF/K}$$

Mismatch: 4.3 %

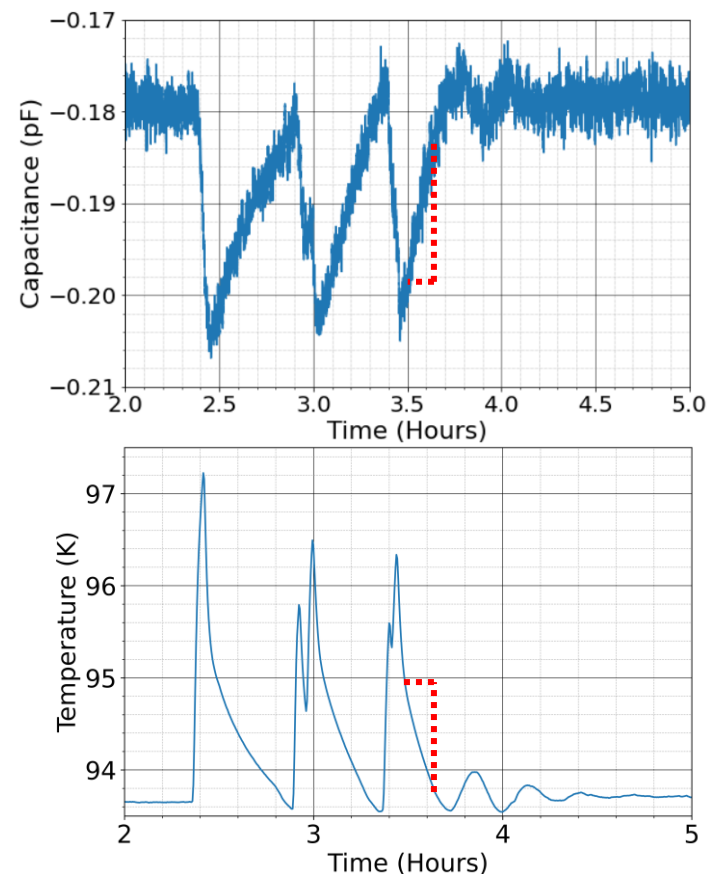
Conclusion:

Unwanted temperature sensitivity is
strongly subdominant to signal from
density changes

Measurement from
recovery after last
mixing heating:

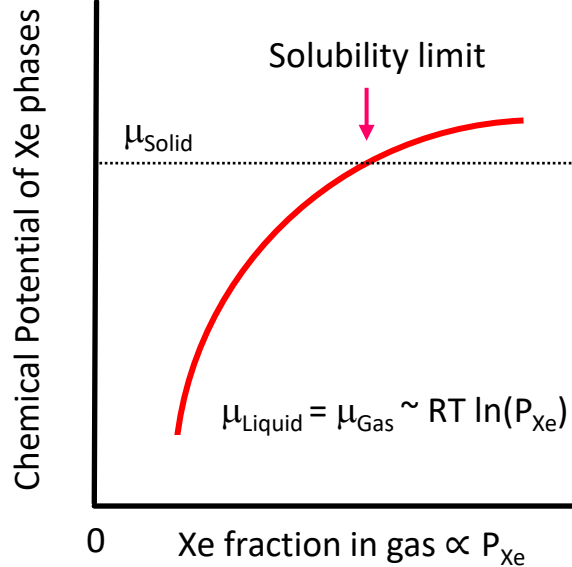
$$\frac{dC}{dT} = -15.46 \text{ fF/K}$$

(Note that we assume
the 2.35% doping has
no effect here.)



Solubility Considerations

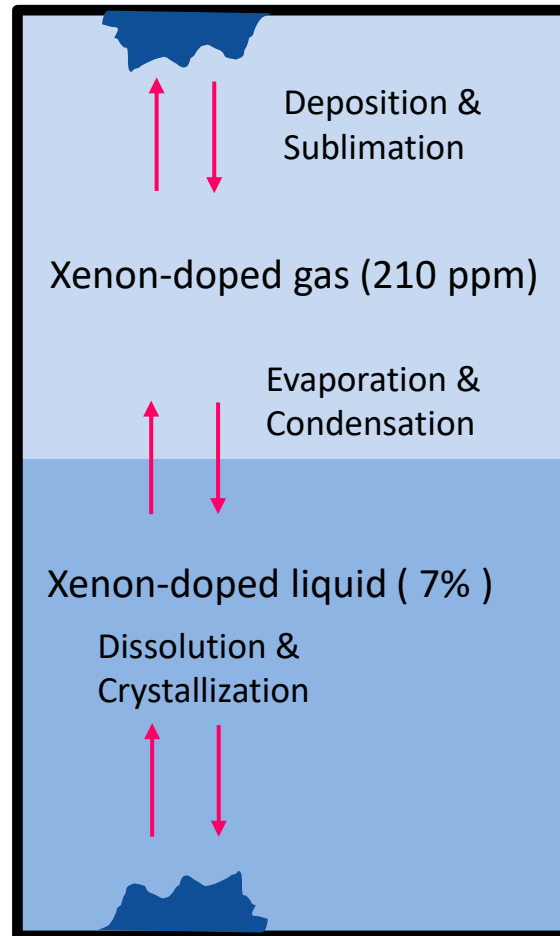
Extrapolating to
 $100/T = 1.054$ from plot at right
 Predicts $n^{\text{Sat}} = 7.1\%$ at 2 bar



$$H = \frac{\text{Xe number fraction in liquid}}{\text{Xe number fraction in gas}}$$

From solubility data we estimate
 $H \sim 250 - 450$ at 2 bar

Strong Distillation Effects!



Detector Vessel

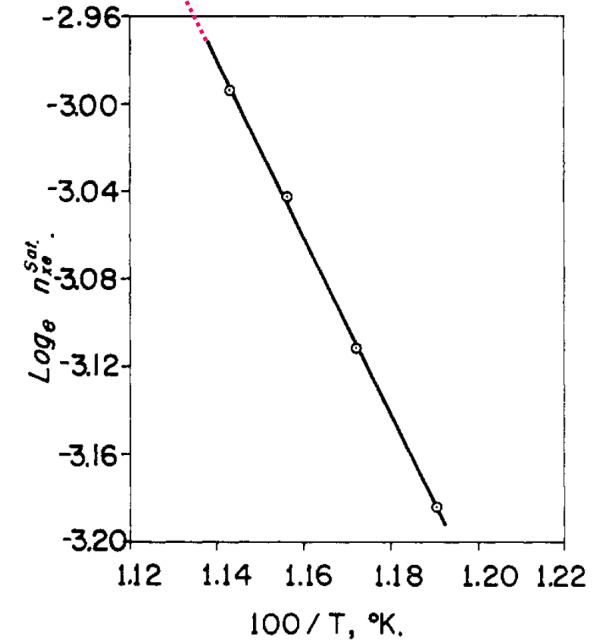


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W. H. Yunker and G. D. Halsey Jr.,
J. Phys. Chem., **64**(4) (1960) 484.