Stabilization of Highly Concentrated Xenon-Doped Argon Mixtures

arXiv:2209.05435

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LIDINE 2022, Warsaw, Poland

September 23rd, 2022

LLNL-PRES- 839848

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under contract DE-AC52-07NA27344. Lawrence Livermore National Security, LLC

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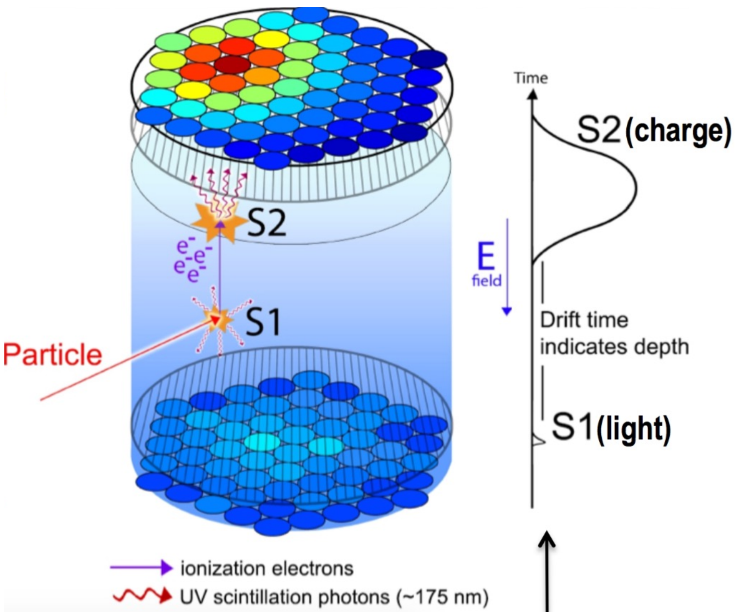
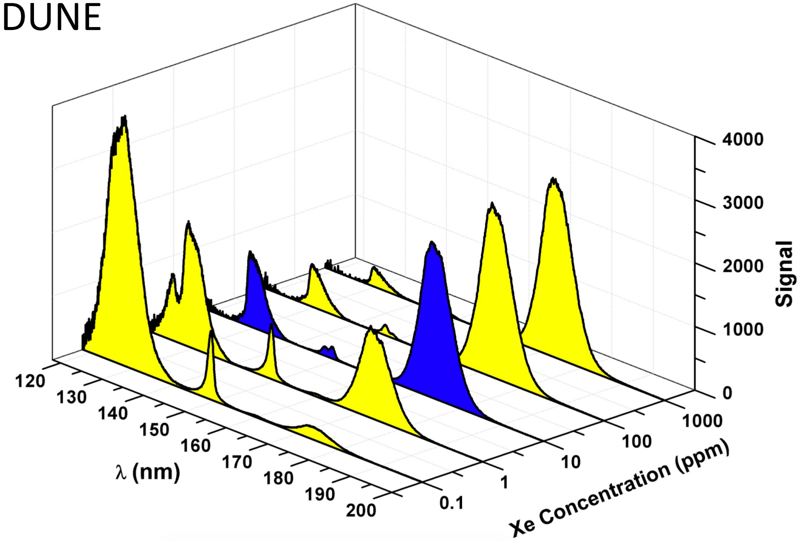
Benefits of xenon doping in liquid argon

- Low-concentration xenon doping in liquid argon is well studied for “wavelength shifting” of argon scintillation light
  - Energy transfer from Ar* to Xe
  - Wavelength shifting of 128nm light, shorter de-excitation time
  - 10ppm xenon in LAr almost completes the “wavelength shifting” process
- This technique may have significant application in large LArTPCs such as DUNE



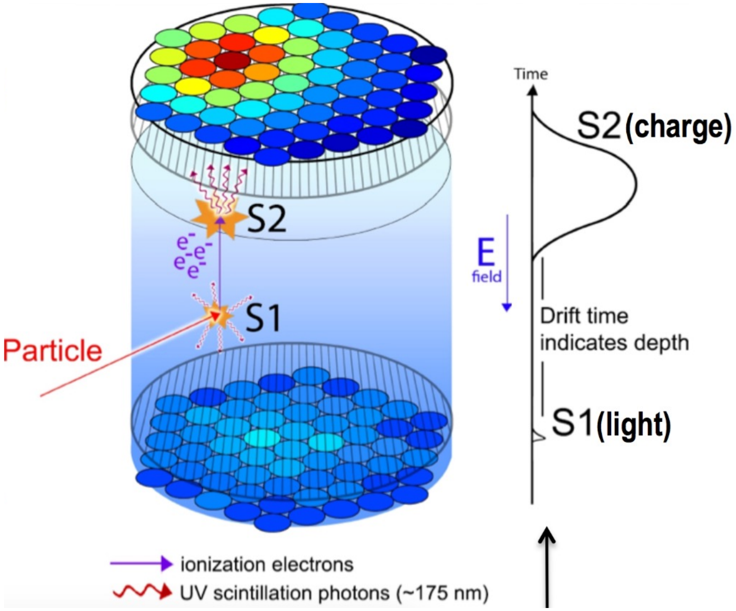


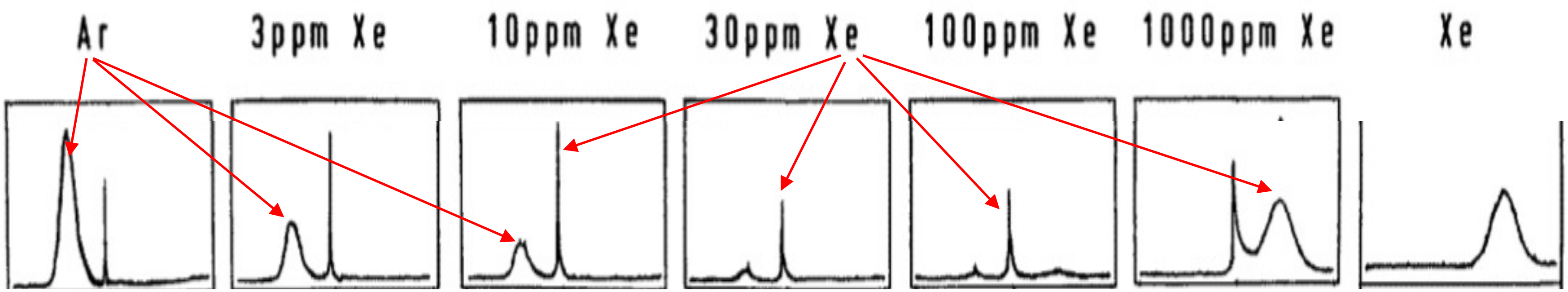
Spectra of scintillation light emission in liquid argon-xenon mixtures
A. Neumeier et al., Europhys. Lett. 109 12001 (2015)

Benefits of xenon doping in liquid argon

Xenon doping enhances electron signal production and detection in a dual-phase argon detector

- Ionization yield in the liquid increases with Xe doping (Penning ionization)
- Electron signals are amplified in the gas phase (lower excitation energy of xenon)
- 10s of ppm xenon in gaseous argon can shift a large fraction of argon electroluminescence to longer (xenon) wavelengths
  - **Higher photon yield**
  - Longer wavelength → **easier detection**
  - Shorter decay → **improved timing**



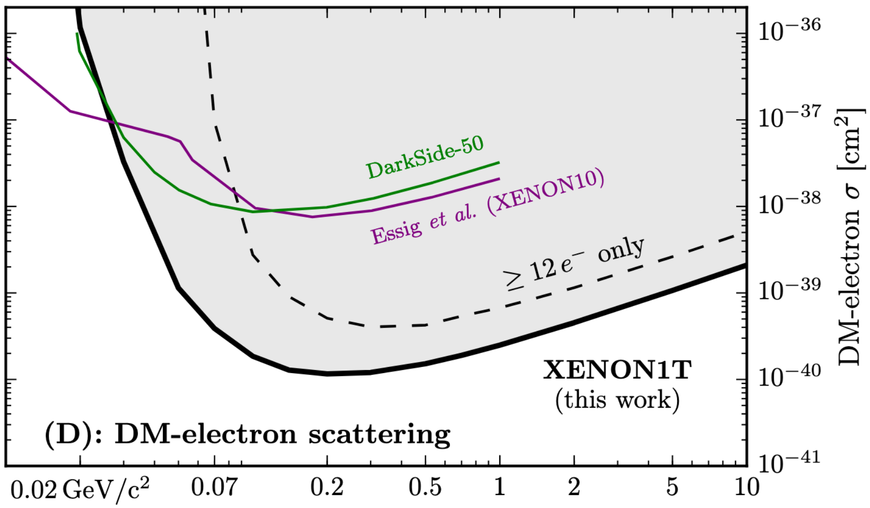


Spectra of scintillation light emission in gaseous xenon-argon mixtures
(T Efthimiopoulos et al 1997 J. Phys. D: Appl. Phys. 30 1746)

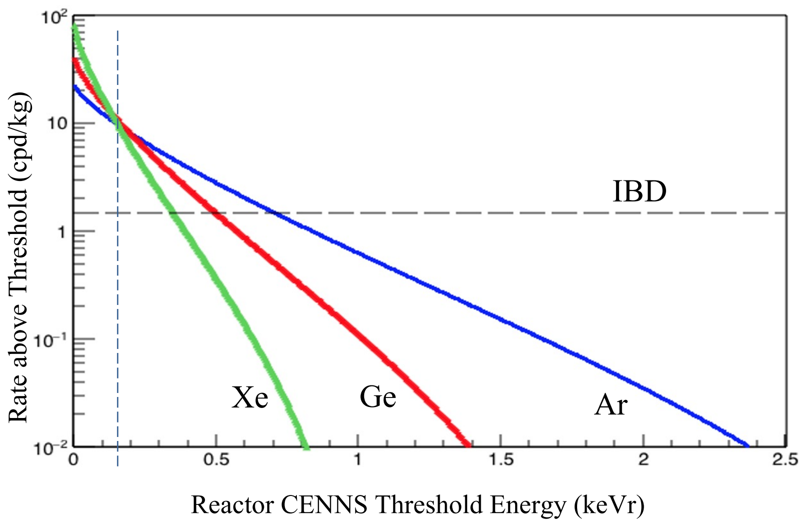
The CHILLAX experiment at LLNL

CHILLAX – CoHerent Ionization Limit in Liquid Argon and Xenon

- **Approach:** dual-phase argon TPC with heavy xenon doping optimized for low-energy S2s
- **Liquid argon target:** *low mass target, low electron background from impurities (colder → less outgassing) and unextracted electron (easier electron extraction)*
- **Xenon-like performance**: *low effective ionization energy, high S2 yield, long S2 wavelength and short S2 decay time*



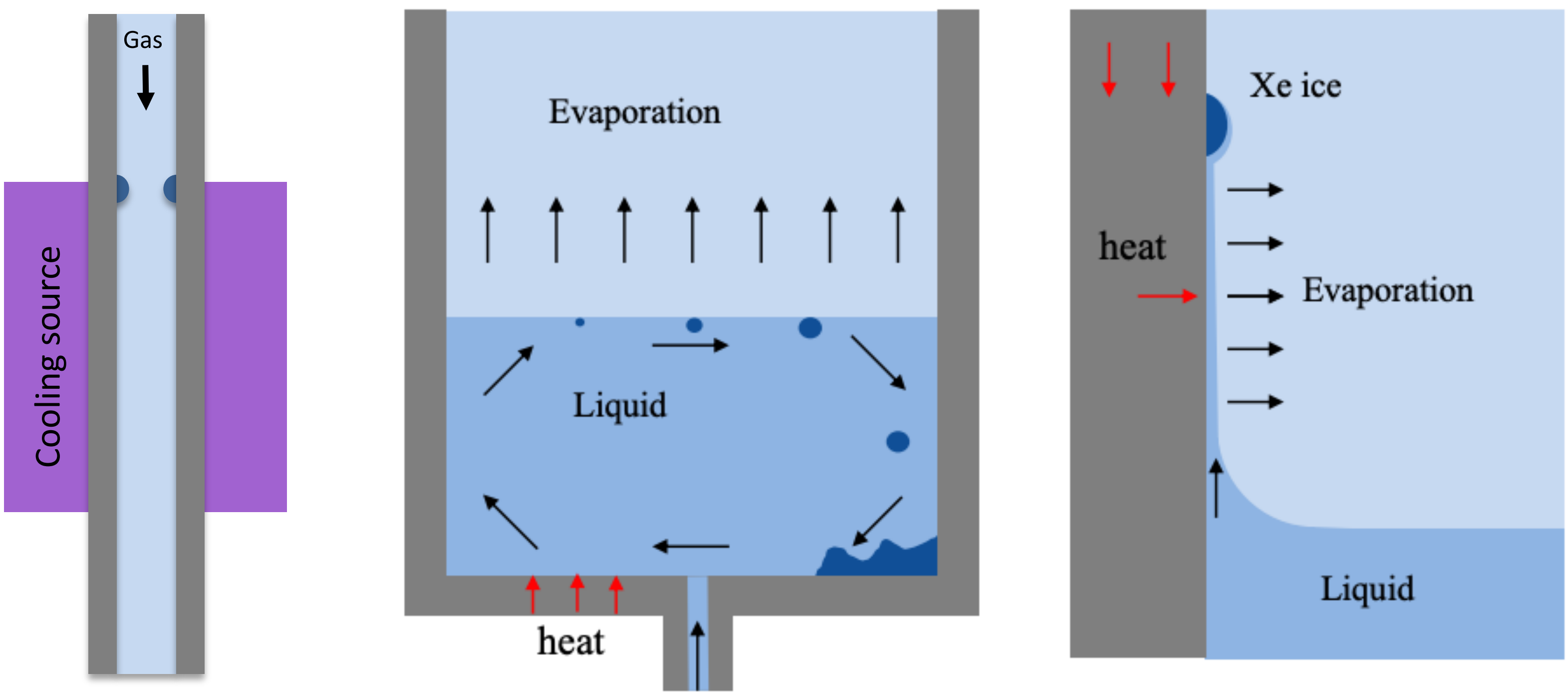
Dark matter-electron scatter limits obtained with Ar/Xe experiments can be further improved, *PRL. 123, 251801*



Estimated CENNS rate in different detector mediums for a reactor of 1GW with 25m standoff

Instability in xenon-doped liquid argon detectors

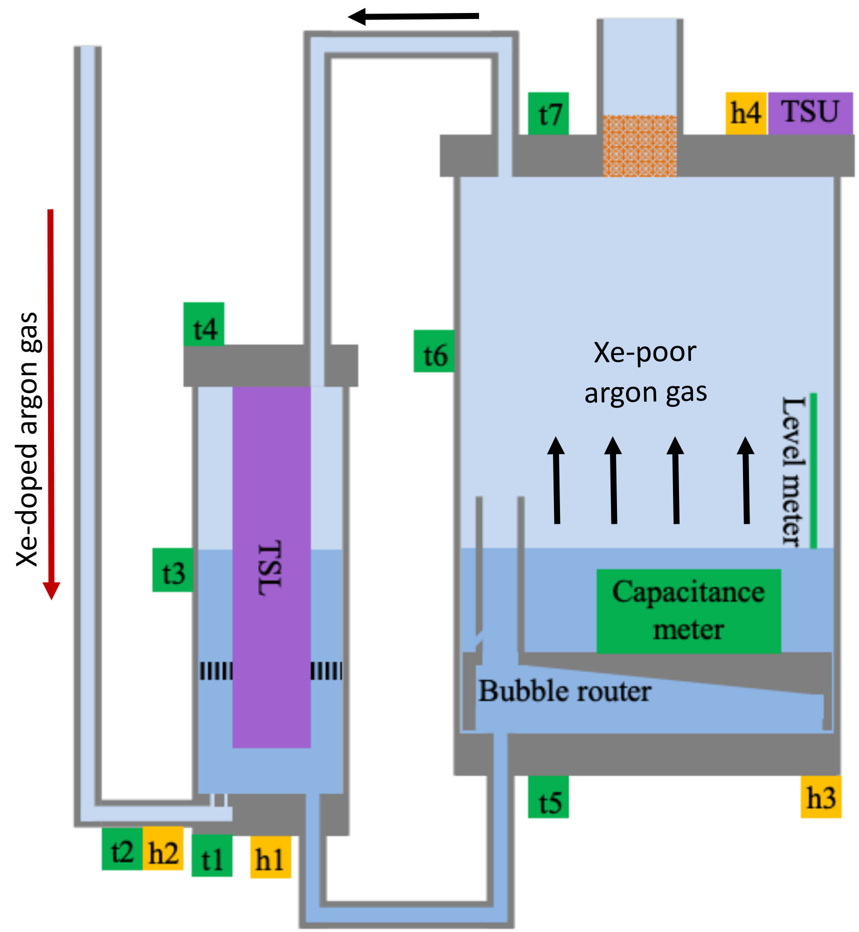
Key challenge with this detector paradigm: instability of xenon doped in argon
Left: Condensation of Xe-rich Ar gas causes Xe to freeze if Xe pressure exceeds saturation vapor pressure
Middle: Evaporation of liquid mixture causes Xe concentration to increase in the liquid
Right: Unintended evaporation of liquid isolated by surface tension can cause Xe ice to form



CHILLAX design

Designed to mitigate instabilities

- Evaporation of liquid mixture from the main detector volume keeps xenon in detector and produces xenon-poor gas
- Xenon-poor gas condenses in HX to produce xenon-poor liquid
- Condensation of xenon-rich doped argon gas (~0.6% xenon by vol) in xenon-poor liquid argon
- Encourage liquid convection in main bath to prevent saturation at the surface
- Control vertical temperature field profile to avoid enhanced evaporation from liquid drawn onto the walls by surface tension
- Xe concentration measured capacitively



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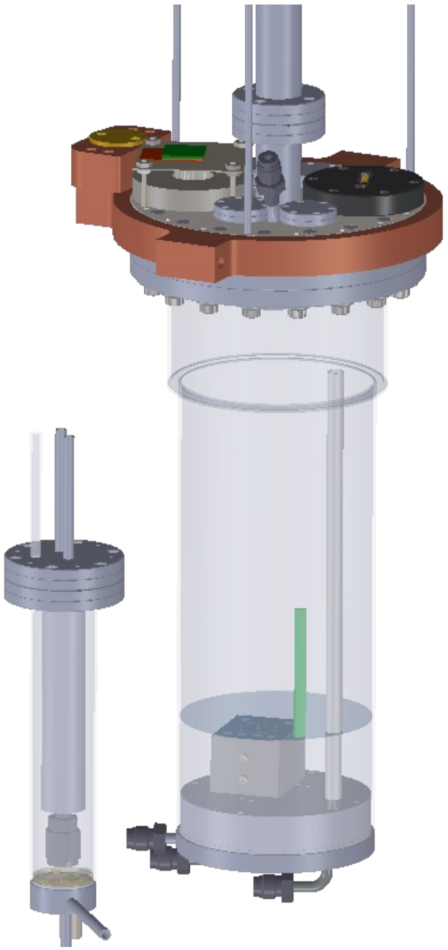
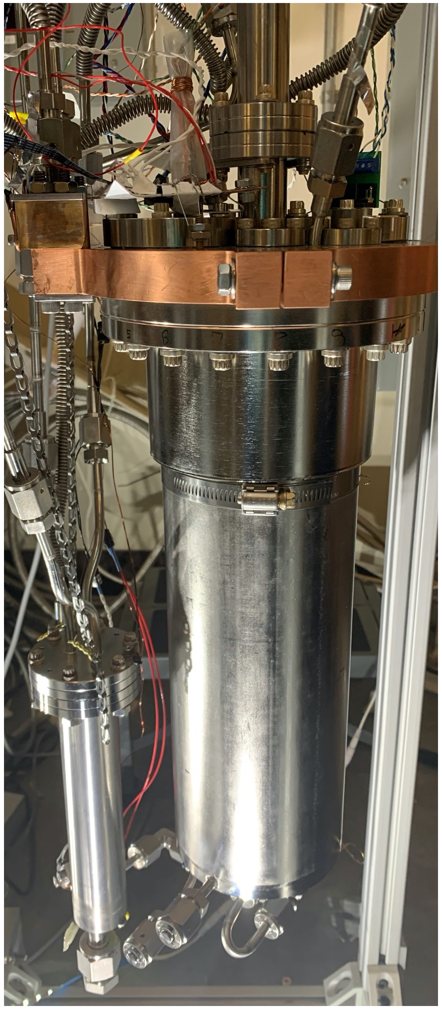
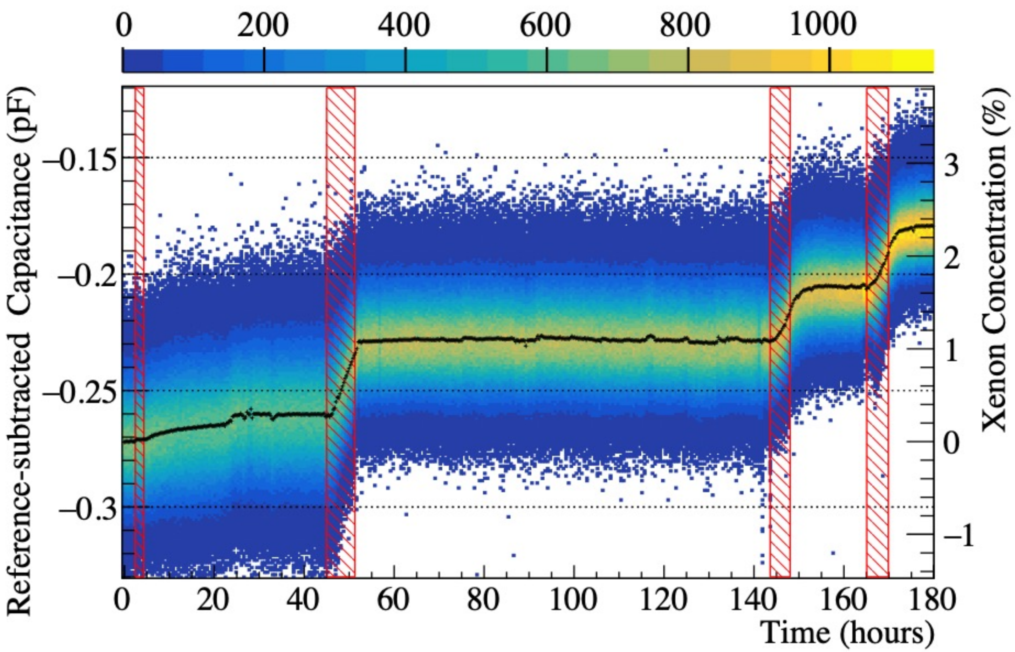




Xenon doping and concentration stability

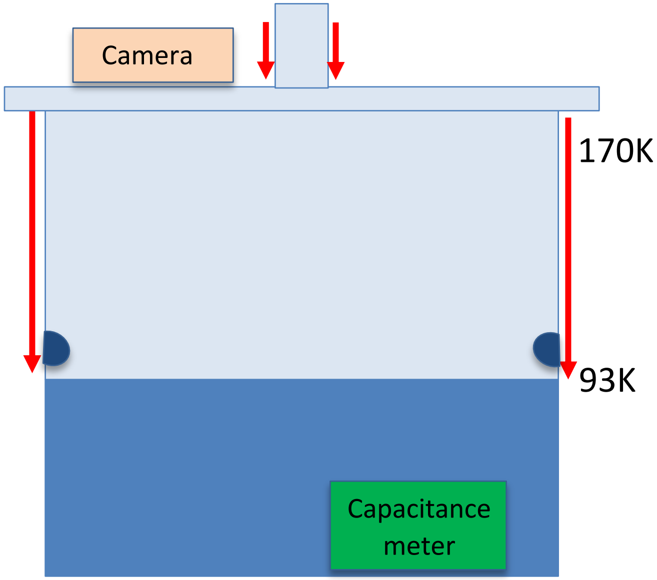
- Xe was introduced into pre-condensed LAr in 4 steps
- System kept stable at 1.8+/-0.005 bar and 93.3K
- 0.6% (mole fraction) of Xe is present in the Ar gas directly condensed
- Xe concentration in liquid measured using a capacitor as liquid dielectric constant changes with xenon doping
- Clear xenon concentration increases observed and maintained during and following doping periods

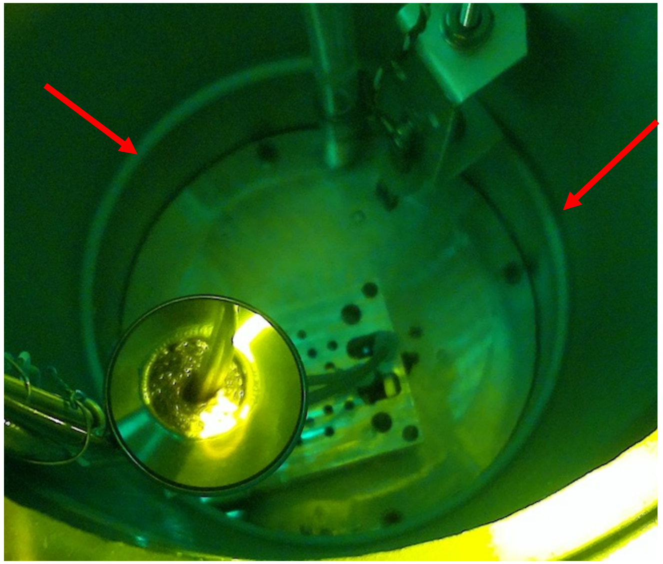


Red hatches: xenon doping periods during CHILLAX operation

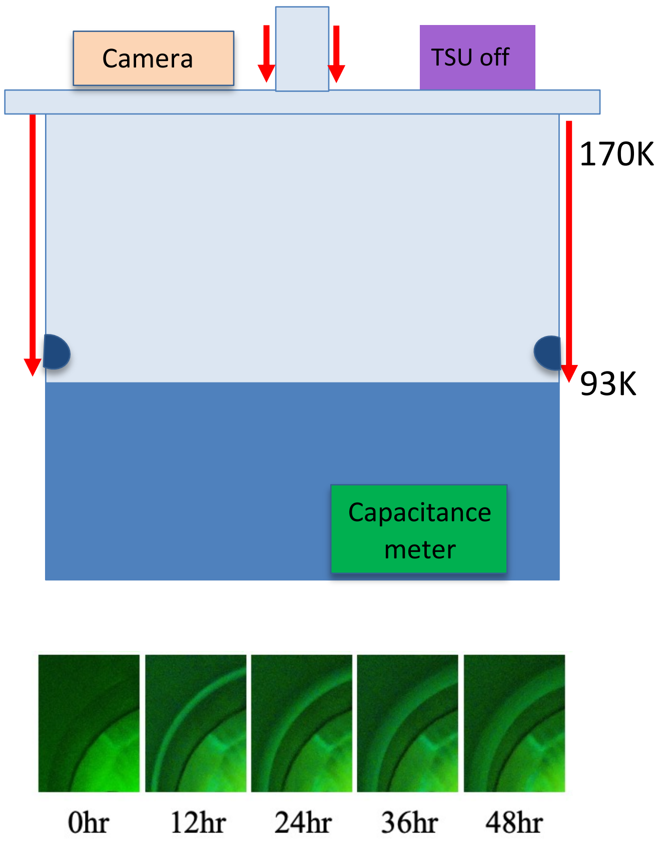
Testing stability of 2.35% Xe-doped LAr

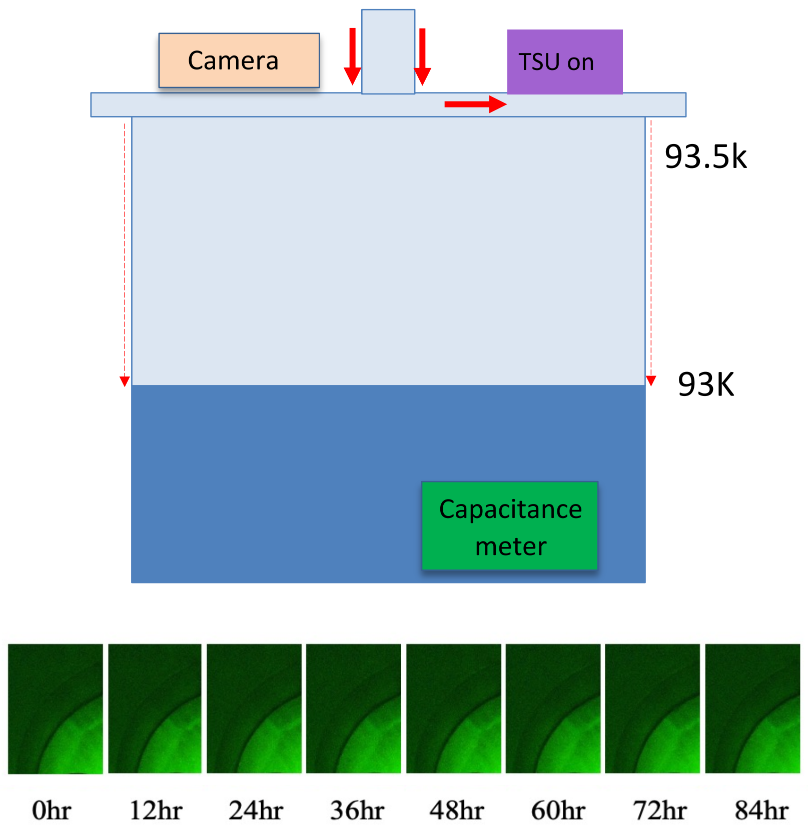
- When detector temperature field was not well controlled, Xenon ice ring visible just above liquid surface
  - Explained as enrichment of xenon in liquid mixture drawn to wall due to surface tension following enhanced evaporation due to heat flowing down the wall





Stability test – Uncontrolled vs. Controlled detector temp. gradient



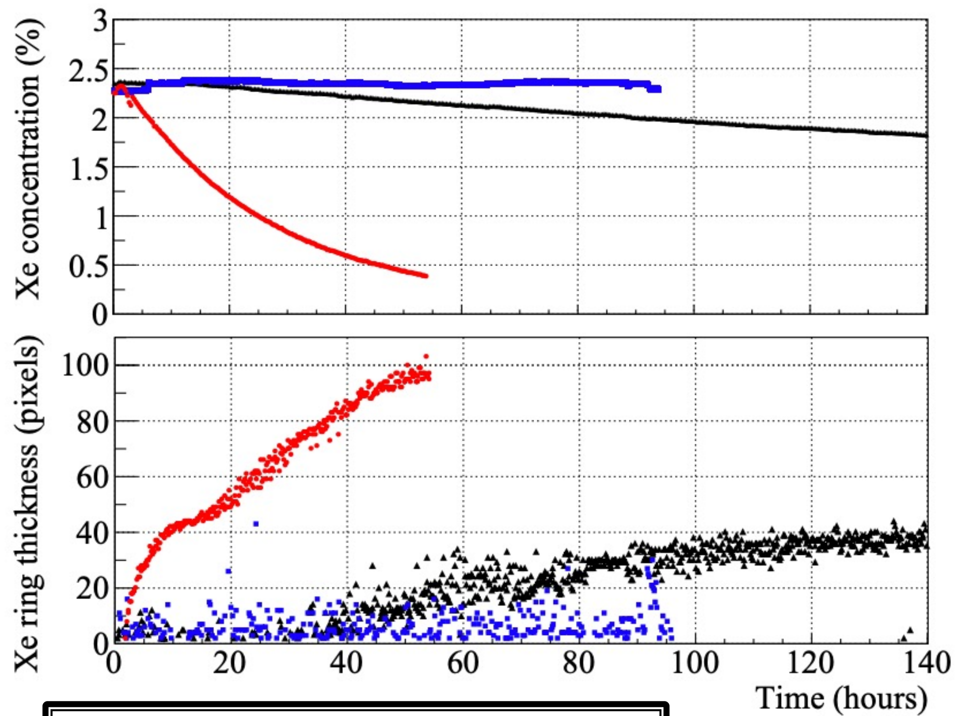


Controlling thermal profile with thermosiphon at top of detector greatly enhances xenon stability in detector volume

Stability test – Rate of xenon ice formation

3 distinct thermal profiles tested

- No degradation in zero gradient test
- Higher rate of xenon precipitation from liquid mixture at larger thermal gradients
- Direct anticorrelation observed between xenon concentration in liquid and thickness of xenon ring above liquid
- Observations indicate having a well-controlled temperature field is critical to ensuring xenon stability in liquid argon



Red: Large thermal gradient above liquid (75 K)
Black: Moderate thermal gradient (10 K)
Blue: Zero thermal gradient

Conclusions

- The benefits to argon detectors through xenon doping are compelling
  - LAr scintillation detectors get "wavelength-shifting" effect from low xenon concentrations
  - LAr ionization detectors can get substantial performance boost with high xenon doping ratios
- Xenon-doped argon detectors can develop instabilities
  - Problems may develop in both heavily and lightly doped systems
  - Doped detectors require specific design elements to mitigate known modes of instability
- The CHILLAX teststand has demonstrated stable doping of xenon into argon up to 2.35% concentration in the liquid phase
  - Cryogenic system is capable of directly condensing 0.6% xenon-rich argon gas
  - System has adequate control to either allow or relieve instabilities
  - Measurements of light yield and charge yield as a function of xenon doping in liquid argon are planned in future tests
- This development has broad impact on future Xe-doped LAr efforts at both low and high concentrations

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Back-up slides

Recoverability of detector state after xenon freezeout

Can the detector's original xenon/argon state be recovered after a xenon freezing event?

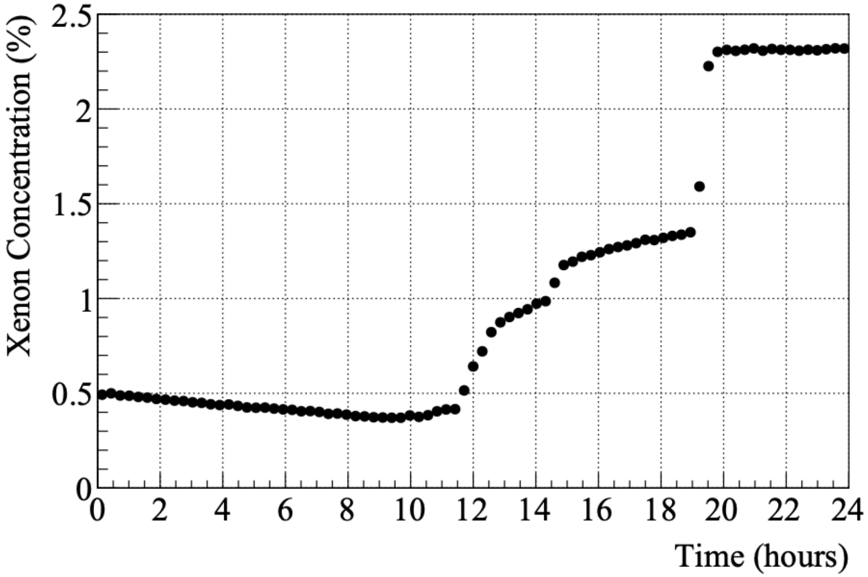
Following test with large thermal gradient on can, turned on TSU to produce Ar condensation on walls

- Xenon ice ring can be melted/recovered into the liquid volume in CHILLAX within ~1 day

Important demonstration for larger scale experiments running on ~years timescale

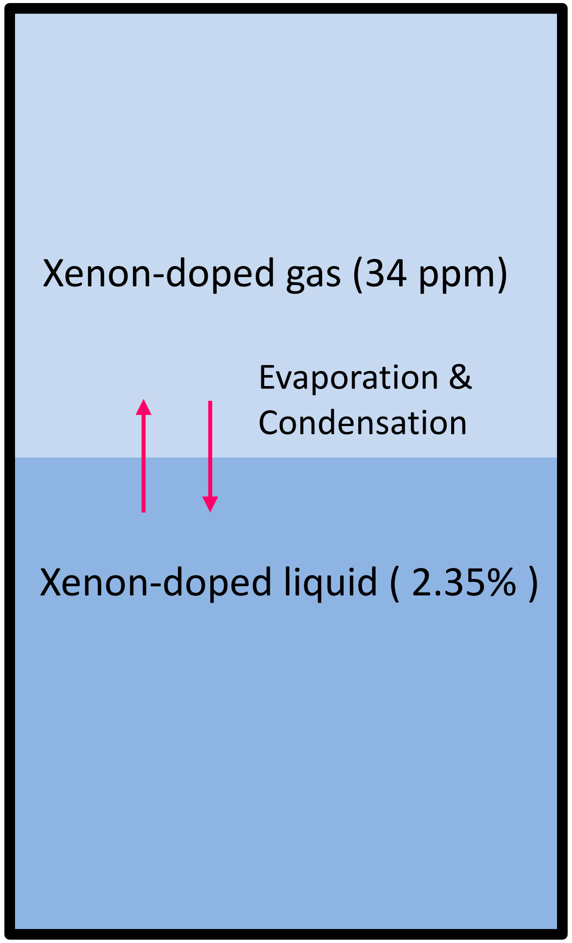
- Detector state recoverable after power outages, equipment failure, etc. could lead to xenon freezing

Liquid xenon concentration in CHILLAX following xenon ice ring formation and reactivation of TSU



Large increases in liquid xenon concentration from ice chunks releasing and melting into liquid volume

Unsaturated mix @ 1.8 bar



Detector Vessel

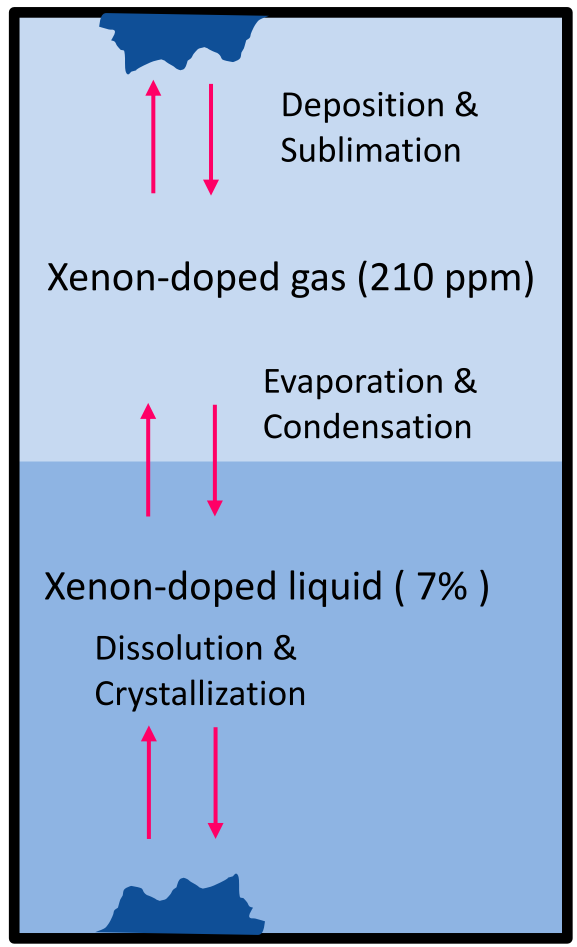
$$H = \frac{Xe\ number\ fraction\ in\ liquid}{Xe\ number\ fraction\ in\ gas}$$

From solubility data we estimate
H ~ 690 at 1.8 bar

Strong Distillation Effects!

Saturated mix @ 2 bar

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



Detector Vessel

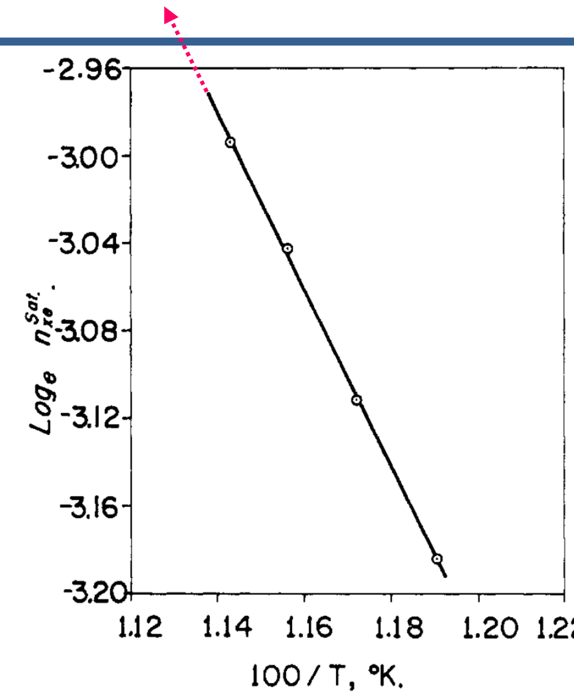


Fig. 2.—Ln n_{xe}^{Sat} *vs.* $100/T$ °K.

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

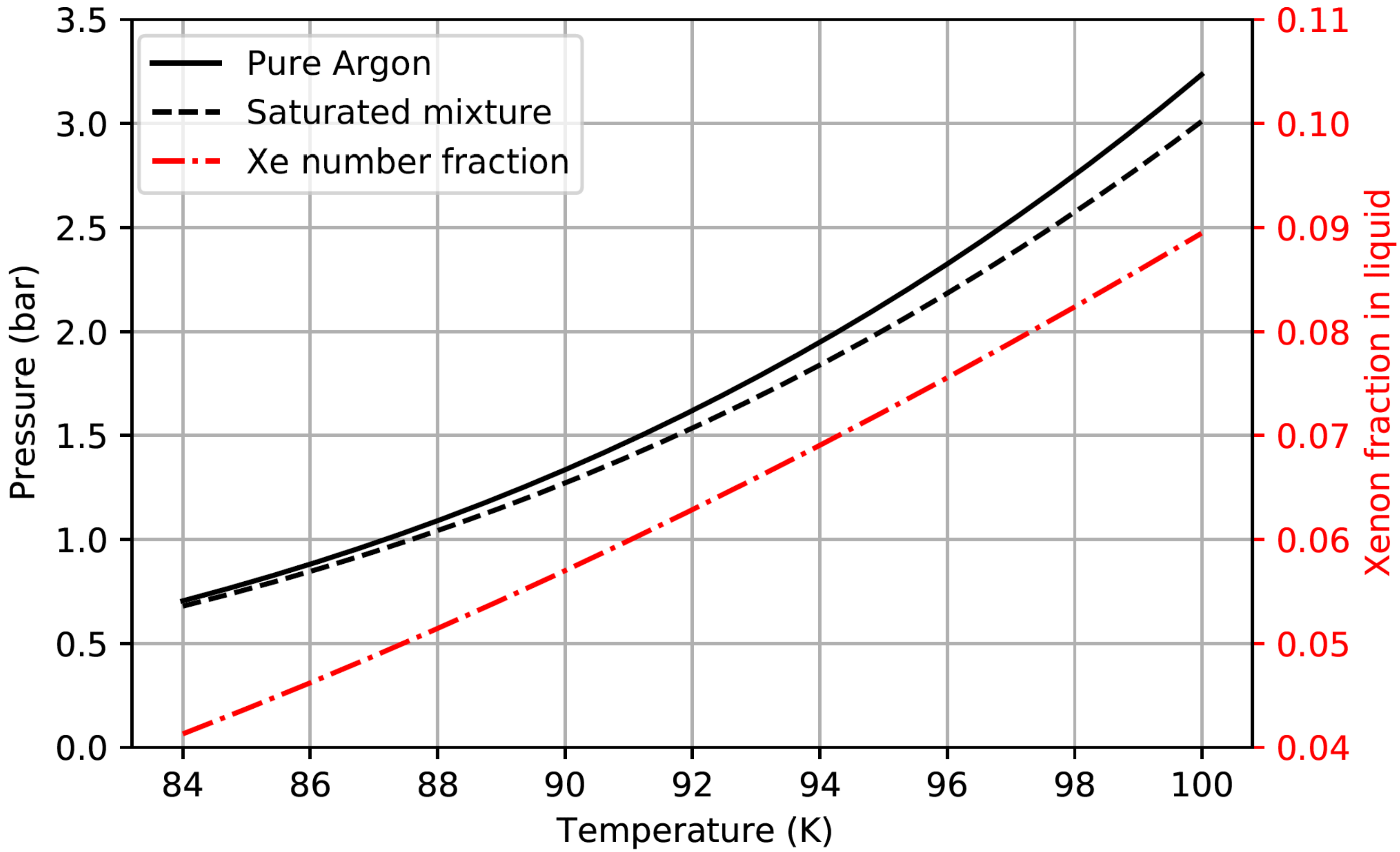
W. H. Yunker and G. D. Halsey Jr., *J. Phys. Chem.*, **64**(4) (1960) 484.

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

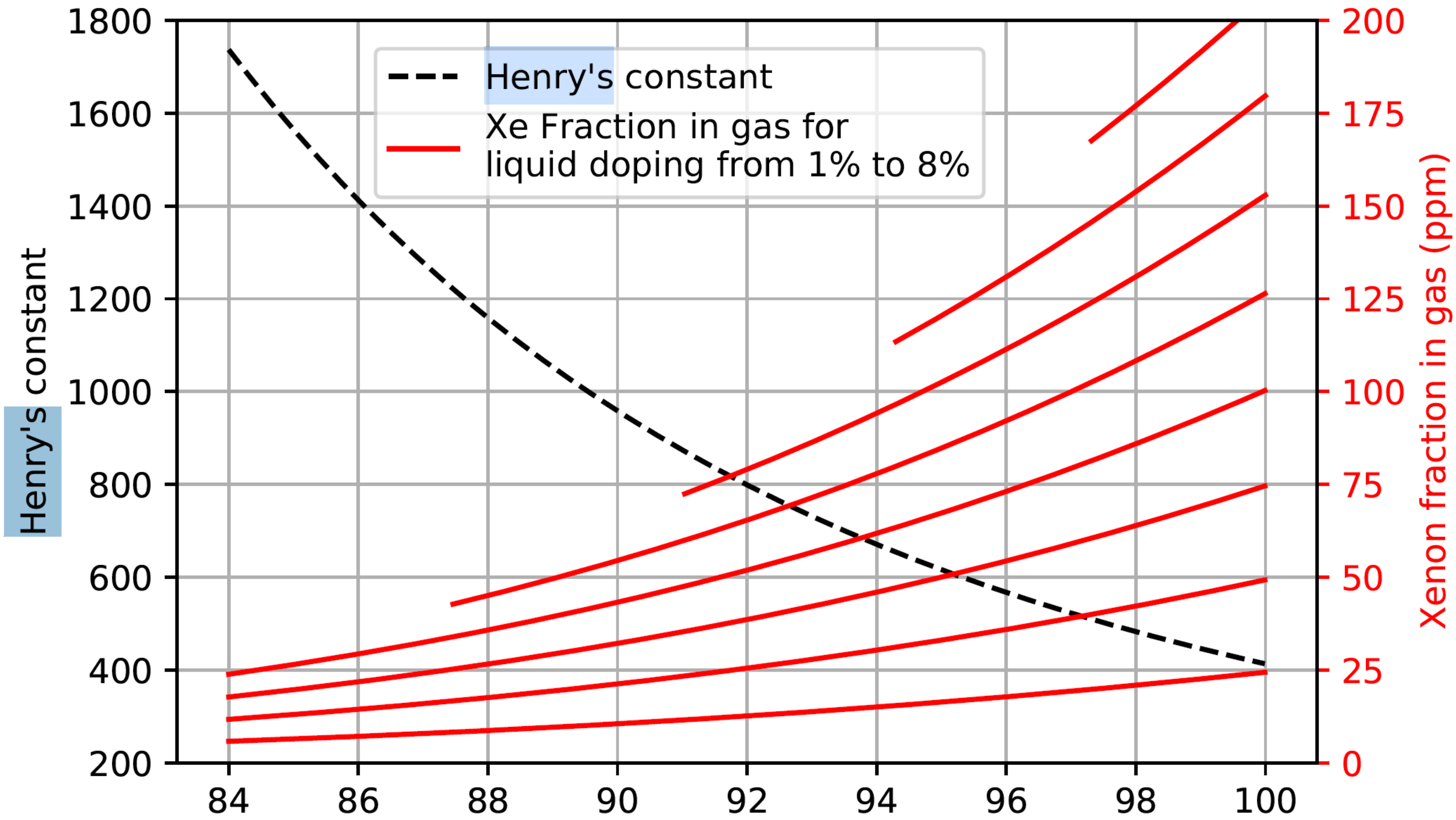
From solubility data we estimate
H ~ 620 at 2 bar

Strong Distillation Effects!

Vapor pressure curves for Xe/Ar mixtures



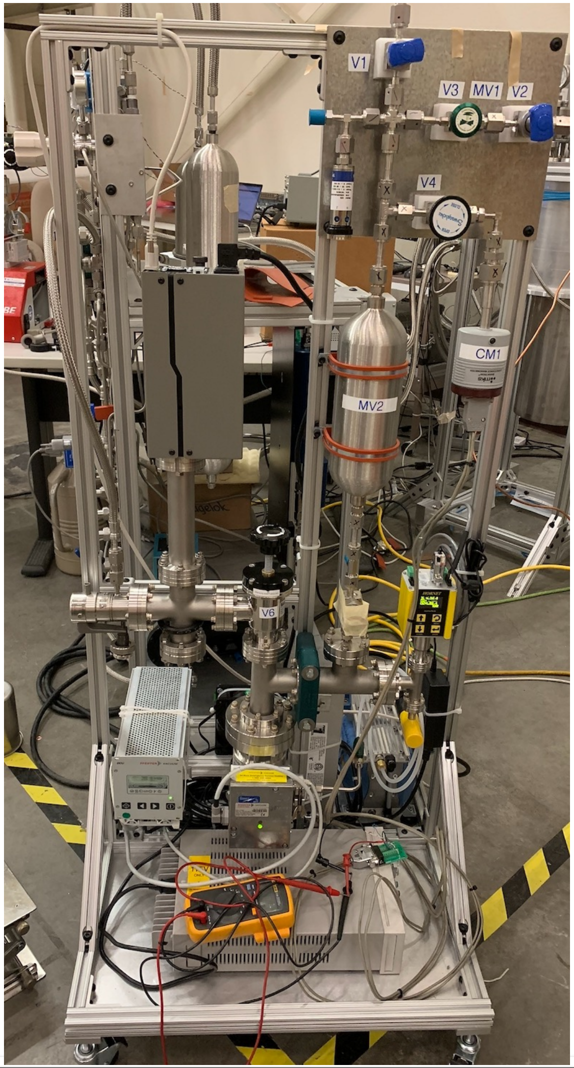
Henry's constant and Xe concentration in gas volume



Xenon concentration measurement in the gas

To gain benefits of xenon doping for electron signal amplification, we need 10s of ppm Xe/Ar in the gas

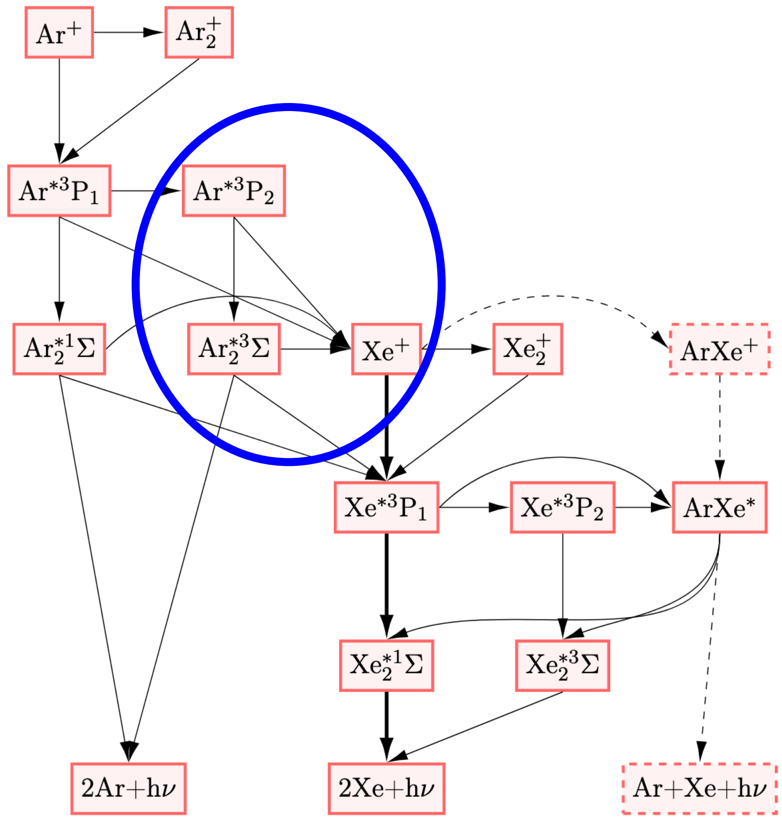
- Goal is to achieve ~30-50ppm Xe/Ar
- Gas composition sampled with RGA200
- Gas dilution through volume expansion, pressure bleeding, pinhole and leak valve control
- Ar-Xe gas mixture in the ppm level can be prescribed for system calibration



Xenon-doping to boost ionization signal?

For detection of small ionization signals, xenon doping may lead to more ionization electrons per energy deposition

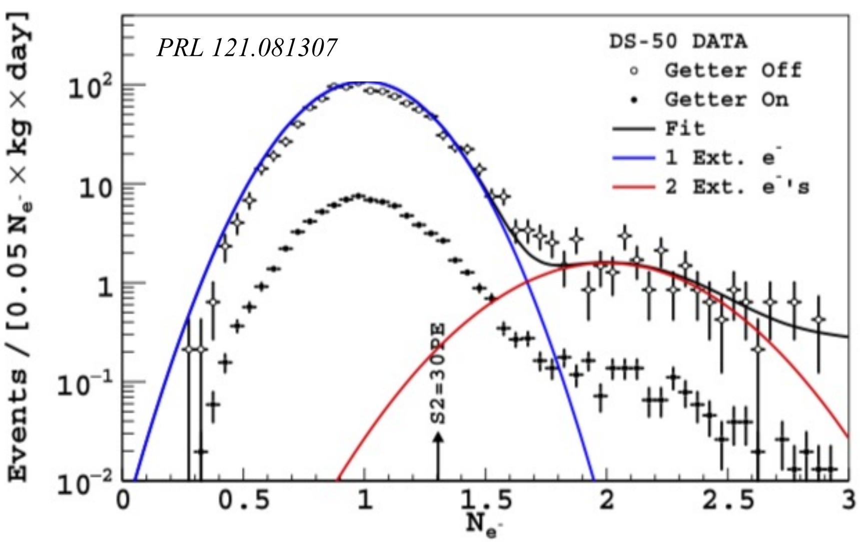
- Some Ar excitation states have higher energy levels than Xe ionization
- Penning ionization of argon excitations could lead to xenon ionization and additional electrons
- May require high electric field to observe significant effect
- Effect to be confirmed experimentally



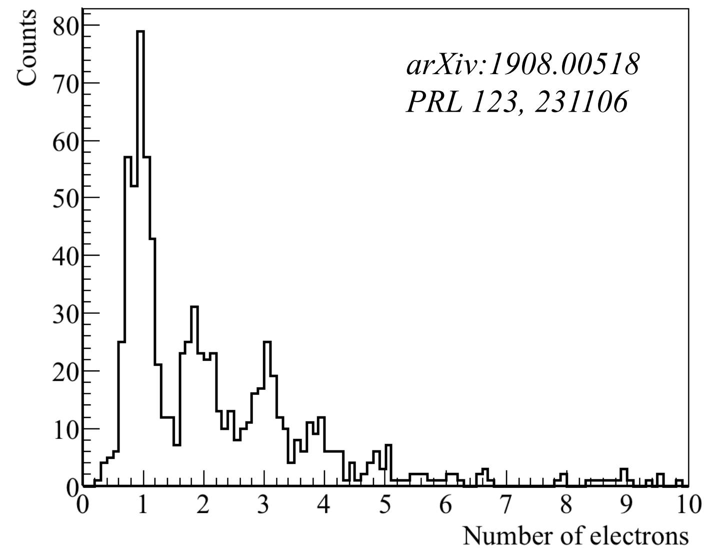
Energy transfer processes in xenon-dopped argon
C. Galbiati et al 2021 JINST 16 P02015

Can one combine advantages of Ar and Xe?

- Argon is a preferred target for kinetic coupling to neutrinos and low-mass DM
- Argon detectors may have a lower ionization background rate
  - Impurity outgassing is a main source for few-electron background (LAr is colder)
  - Unextracted electrons from liquid into gas is another (reduced in LAr)
- Xenon detectors have outstanding energy resolution in the few-electron region
  - Longer wavelength light reduces complexity of signal collection



Single and double electron spectrum in DarkSide50

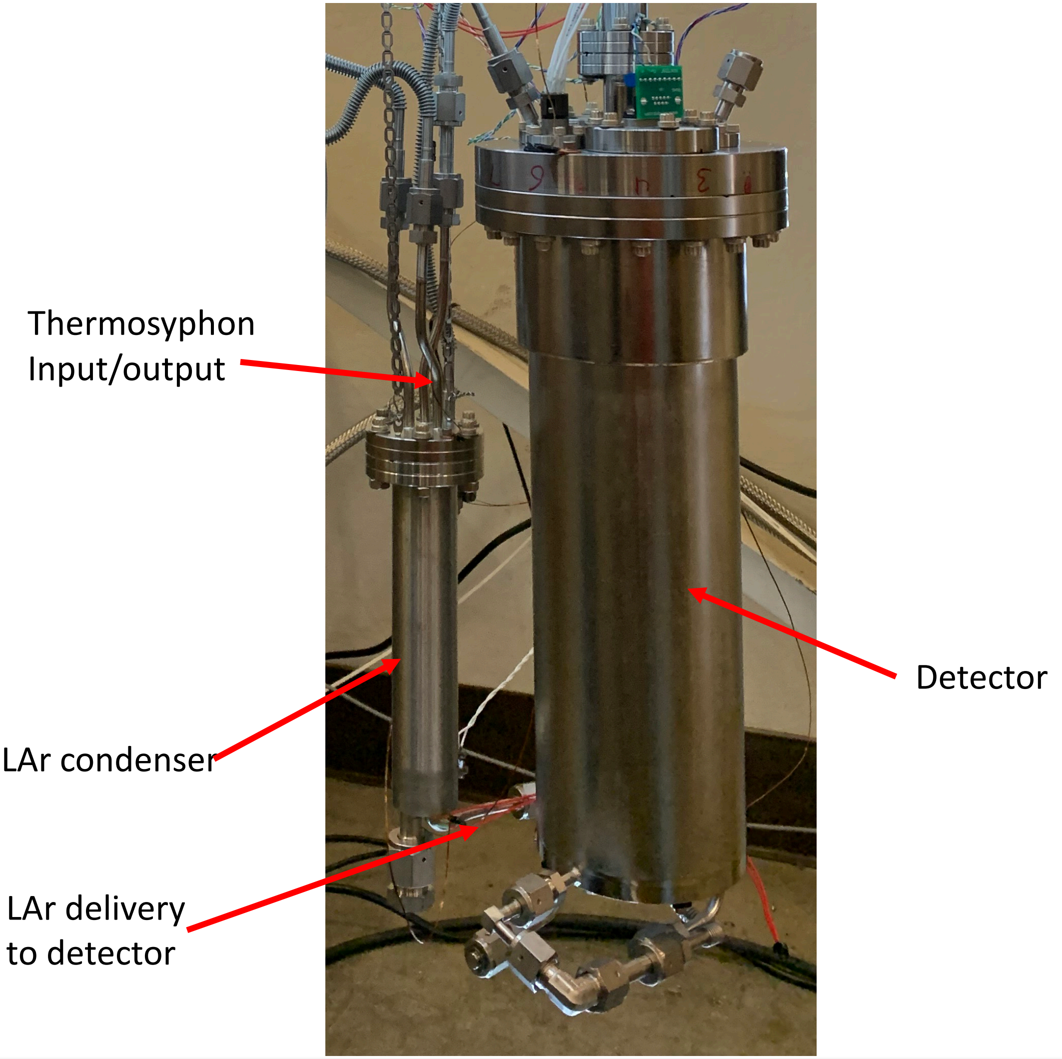


Few-electron spectrum (400eV Xe recoils) in LLNL XeTPC

Xenon delivery in CHILLAX

To obtain %-level xenon doping, CHILLAX adopts a different condensation scheme

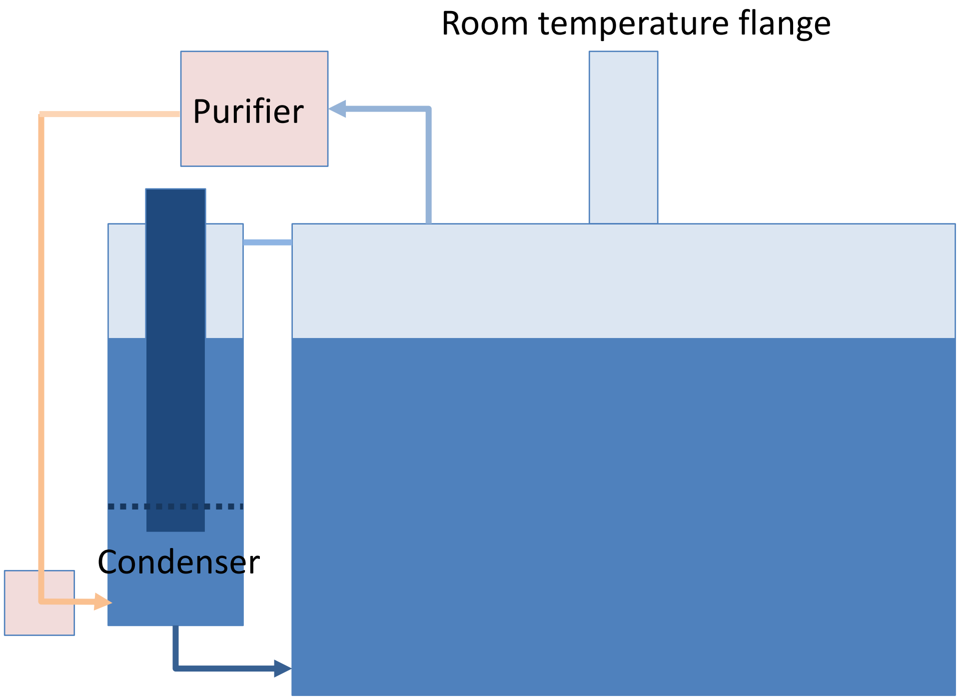
- Introduce 1-2% Xe-Ar mixture gas directly into liquid argon
- Full xenon capture in liquid (ice can dissolve if it forms)
- Direct cooling applied to liquid argon (through thermosyphon) next to detector
- Pre-heating Xe-Ar gas before entrance to condenser



Circulation-purification in CHILLAX

Continuous circulation and purification are needed to keep the target clean

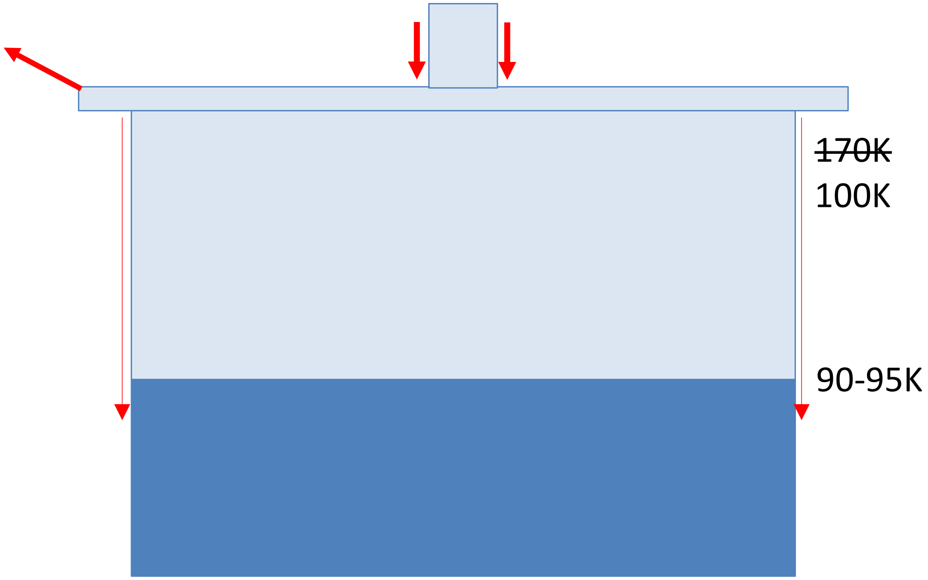
- Ideally, we would move liquid through purifier
  - No phase transition/distillation
  - No xenon congregation
- We are currently testing circulating gas from detector volume
  - No xenon congregation outside detector argon volume
  - Liquid convection could prevent xenon ice forming



Retaining xenon in liquid mixture

We added a thermal link to the cryocooler to intercept heat leak down the support

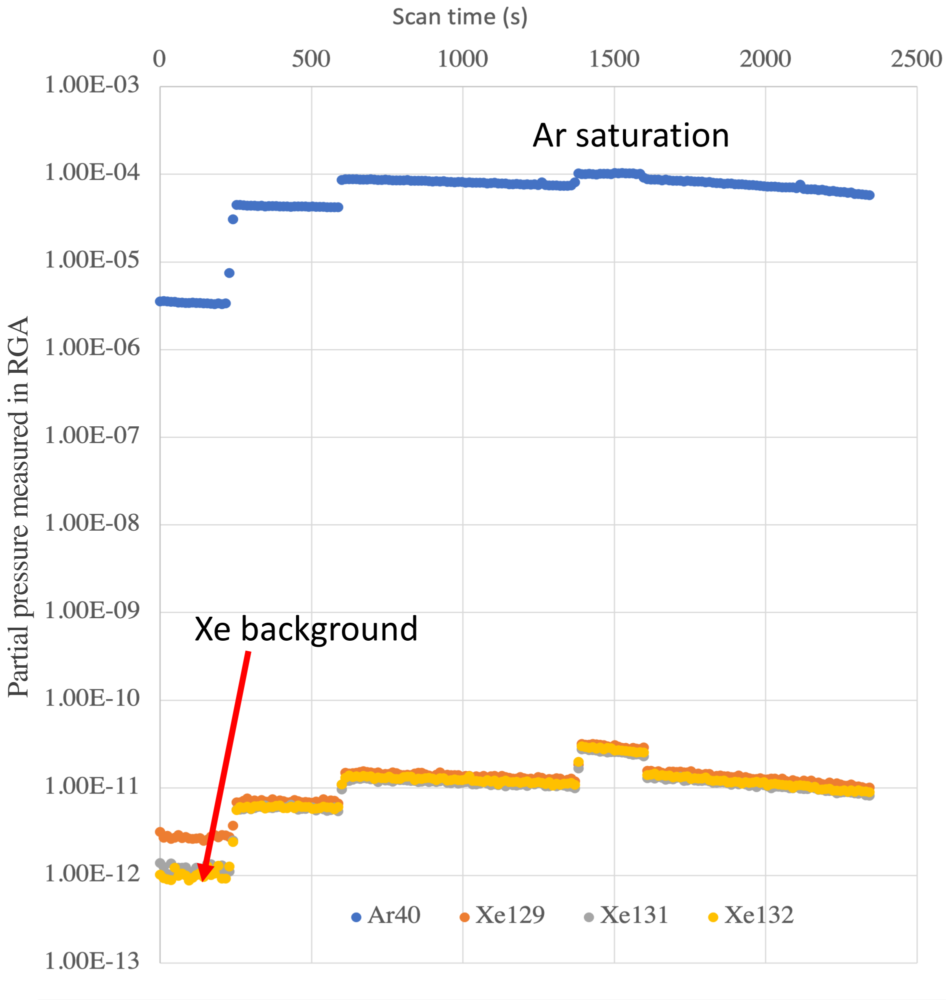
- Detector flange cooled to ~100K while liquid mixture stays at 90-95K
- Xenon ice forming is much slowed
- Will add thermosyphon for stronger cooling (possibly produce Ar reflux on walls)



CHILLAX Sampling system calibration

RGA scan of prescribed Ar-Xe mixture

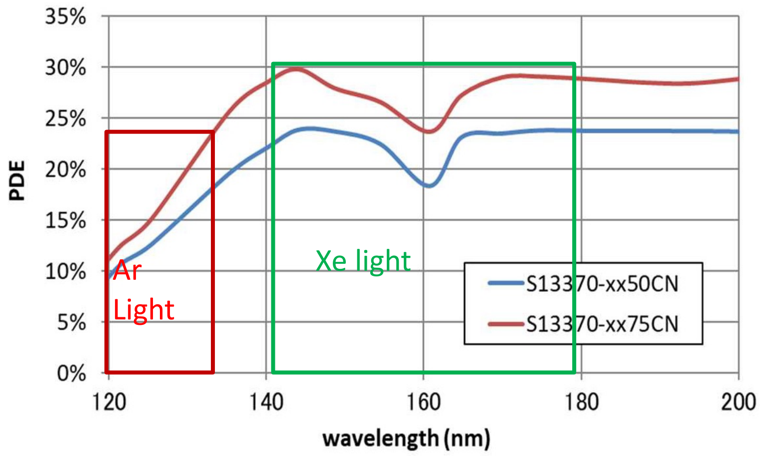
- Sample contains 2ppm Xe/Ar
- Scan used both FC and CEM
- RGA measured ~0.6 ppm
- Complete calibration curve under development
- We observed gaseous xenon concentration in the 10s of ppm region for liquid doping of ~1-2%
- High voltage system to measure electroluminescence gain in preparation

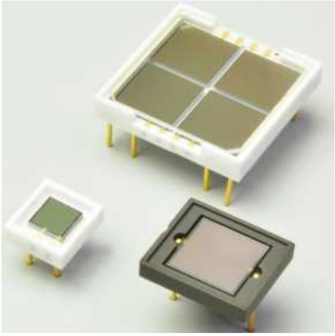


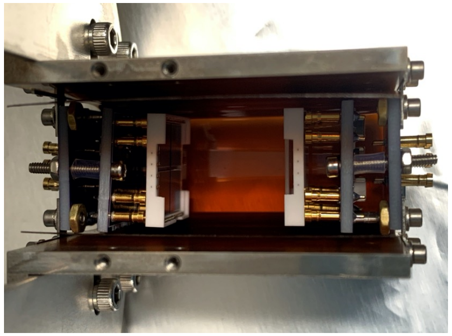
CHILLAX detection of VUV photons

At ~10s of ppm Xe/Ar ratio in the gas, electroluminescence signal may contain Ar^*_2, $ArXe^*$ and possible Xe^*_2 light

- Light detection system needs sensitivity to 128, 147 and 175nm light
- Hamamatsu VUV4 SiPMs are chosen based on their specs
- In-house, onboard, cold amplifiers were built for two types of VUV4 SiPMs (with quartz window and without)
- SiPM modules deployed in CHILLAX (both pure argon and argon-xenon mixture)



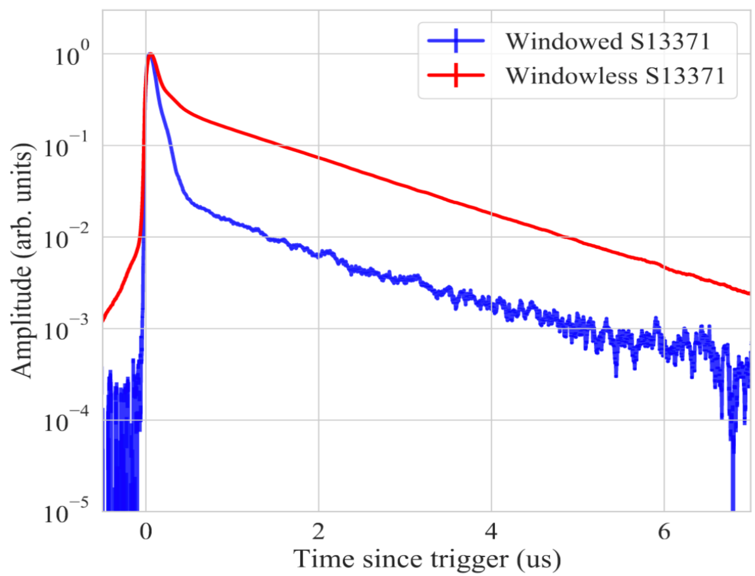


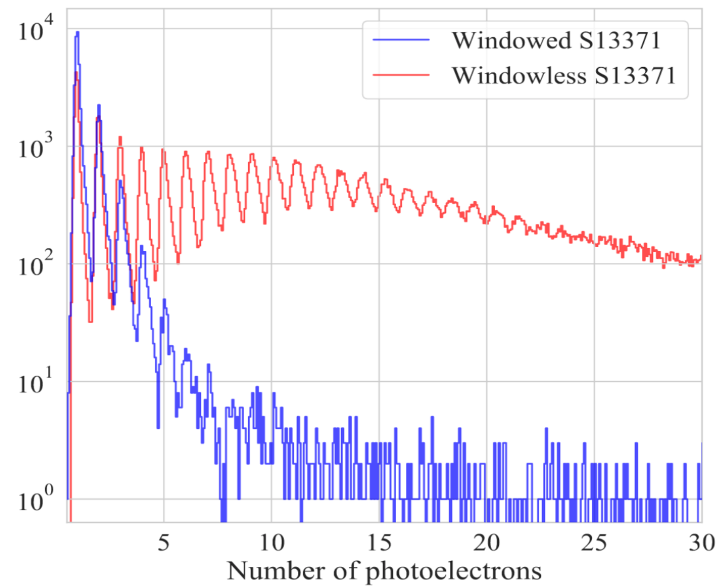


Direct detection of 128nm light

We designed control experiments using VUV4 SiPMs with and without quartz windows (160nm cutoff) to verify direct UV sensitivity

- VUV4 SiPMs with quartz window detected < 1% of light compared to windowless ones
- VUV4 SiPMs with quartz window observed weak argon triplet scintillation component



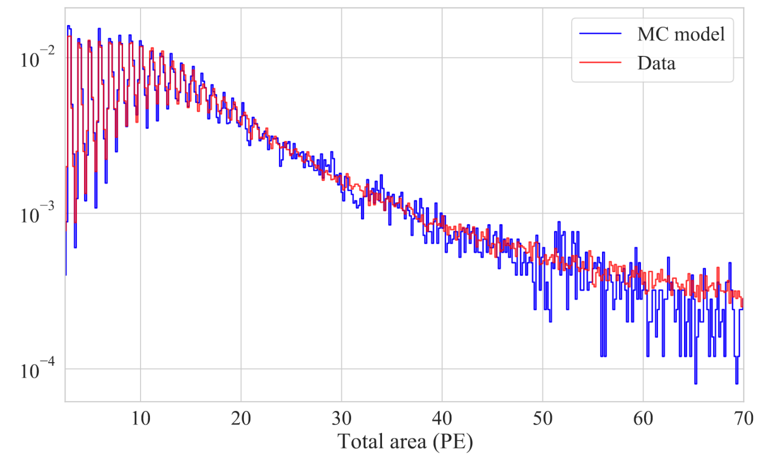


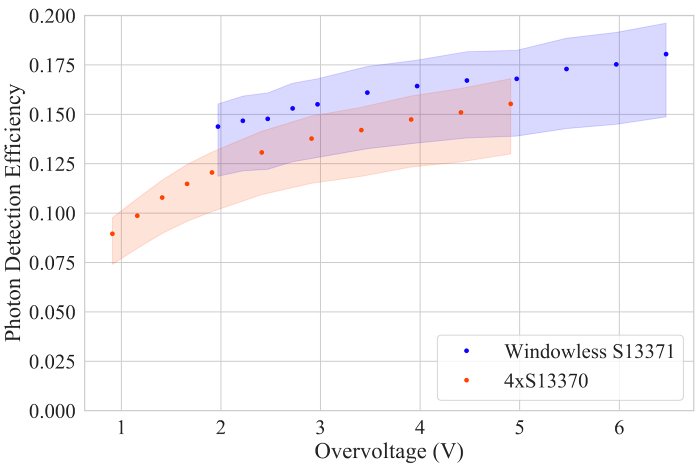
Average SiPM waveforms (left) and energy spectra measured with windowed (blue) and windowless (red) VUV SiPMs for 241Am radiation in pure liquid argon, confirming 128nm sensitivity

Detection efficiency for 128nm light

We measured appreciable PDE values for VUV4 SiPMs at 128nm

- MC simulation tracks data well
- ~15% PDE at 128nm
- Previous characterizations of VUV4 SiPMs at 175nm reported lower than quoted PDE (possibly due to quartz window)
- VUV4 SiPMs are suitable for direct liquid argon light detection!
- Publication arXiv:2202.02977, accepted by JINST





Fitted SiPM spectra for pure argon scintillation (top) and the evaluated SiPM PDE (bottom)

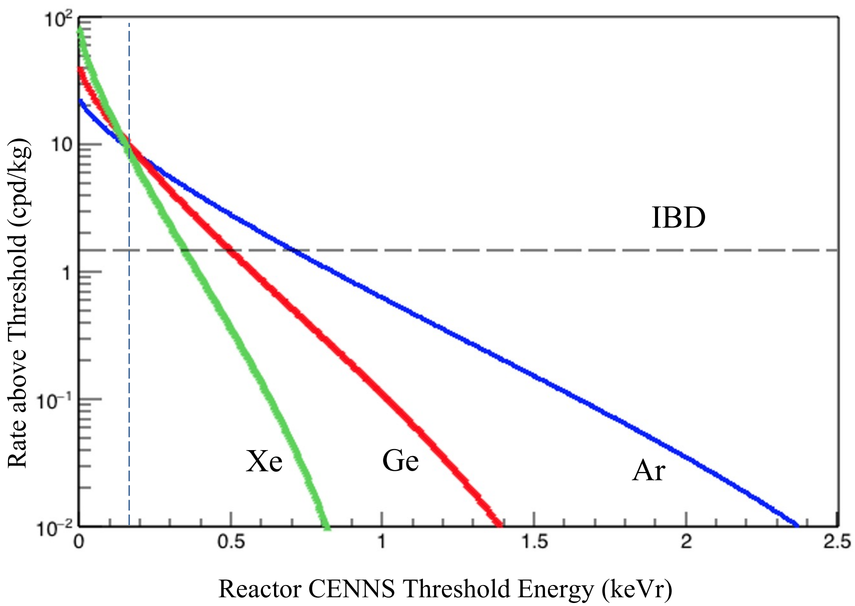
CEvNS rate: foundation for science & application

A CEvNS experiment will first need sufficient statistics to study science or applications

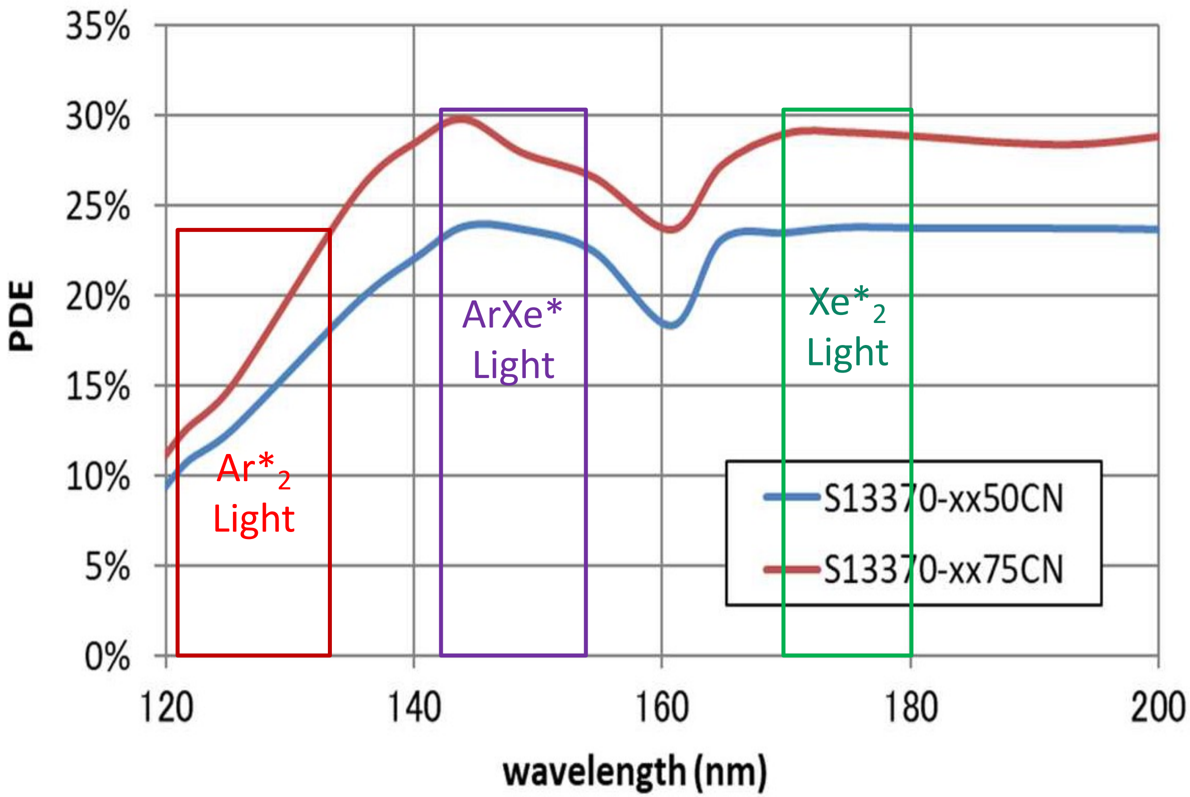
- A low (enough) energy threshold
- A big target mass
- A low background rate

For the case of reactor CEvNS detection

- Signal rates in Ar/Xe/Ge/etc are comparable at a few hundred eV energy threshold
- A zero-threshold Ar(Ge) experiment will see 2(5) time more events per unit mass
- Large target mass and low background have been demonstrated in Ar/Xe detectors



Estimated CENNS rate in different detector medium for a reactor of 1GW with 25m standoff


35%
30%
25%
20%
15%
10%
5%
0%
PDE
120
140
160
180
200
wavelength (nm)
Ar*2 Light
ArXe* Light
Xe*2 Light
S13370-xx50CN
S13370-xx75CN