

# Measuring cosmogenic activation rates in active detector material

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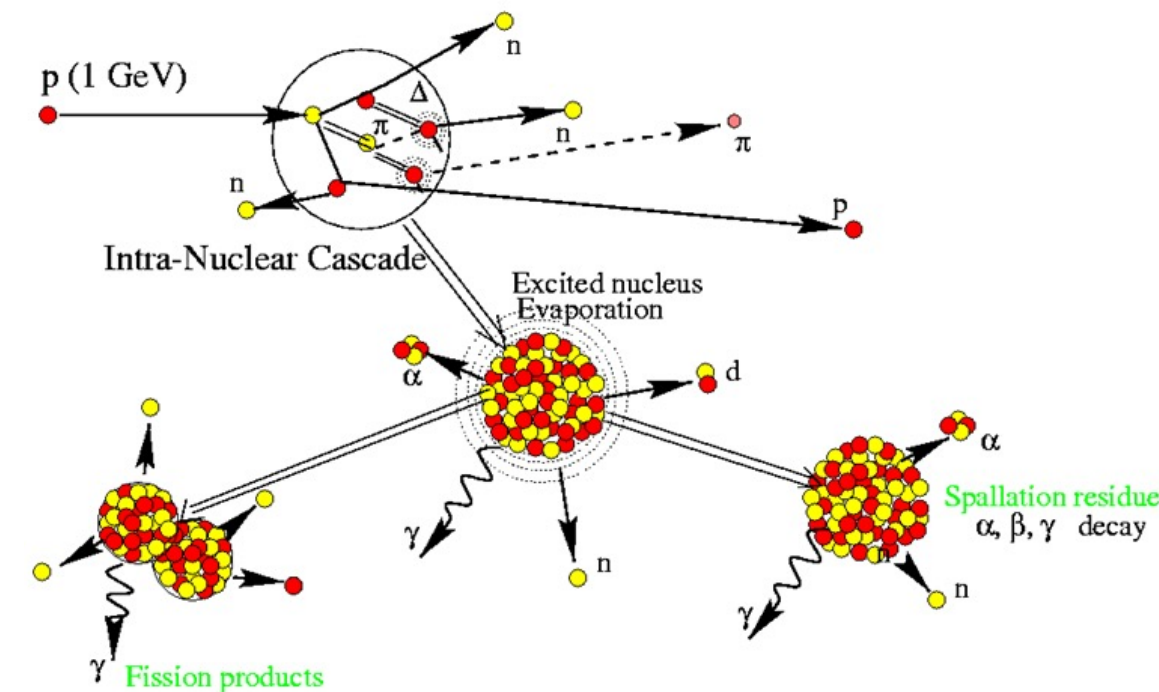
**CPAD Instrumentation Frontier Workshop 2021**

**Virtual Event @ Stony Brook University**



# Cosmogenic Activation

- Radioactive isotopes produced by cosmogenic particle interactions in detector materials can be one of the leading sources of backgrounds in rare event searches
- Understanding the production rate of these isotopes is extremely important in order to evaluate the total surface residency time, transportation options, and storage requirements for low background detector components
- Small production rates and low energy decays of interest for next generation dark matter experiments (tritium,  $^{39}\text{Ar}$ ) make it difficult to measure sea-level activation without building a full-scale experiment



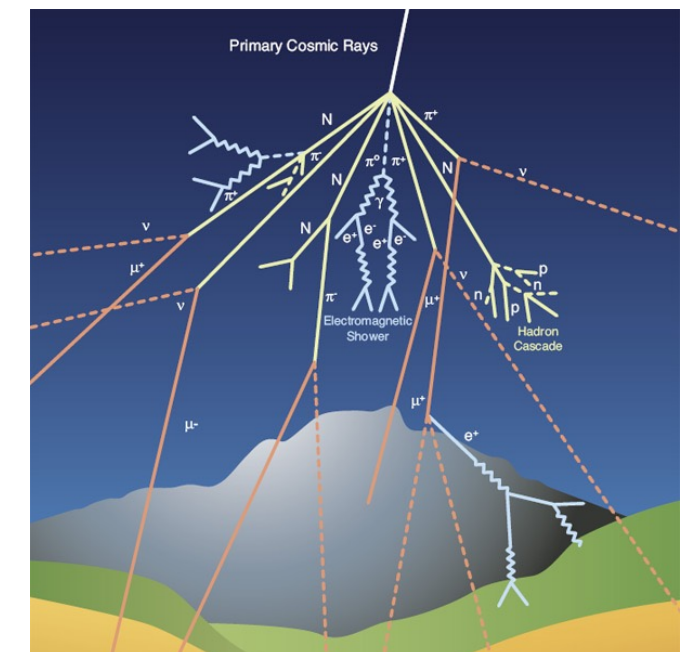
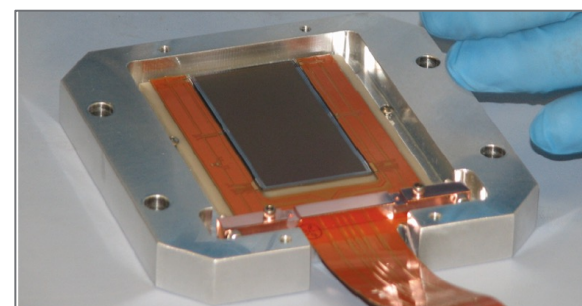
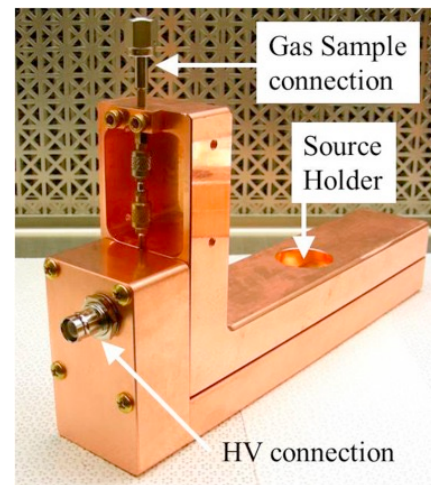
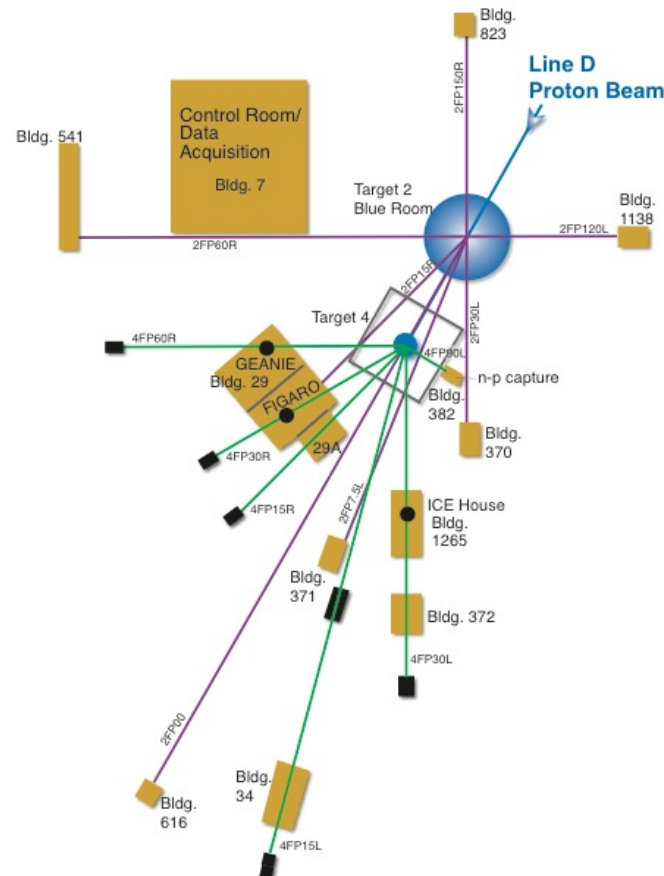


# Measurement Technique

Use high intensity neutron beam to greatly increase production rate compared to sea-level cosmic rays

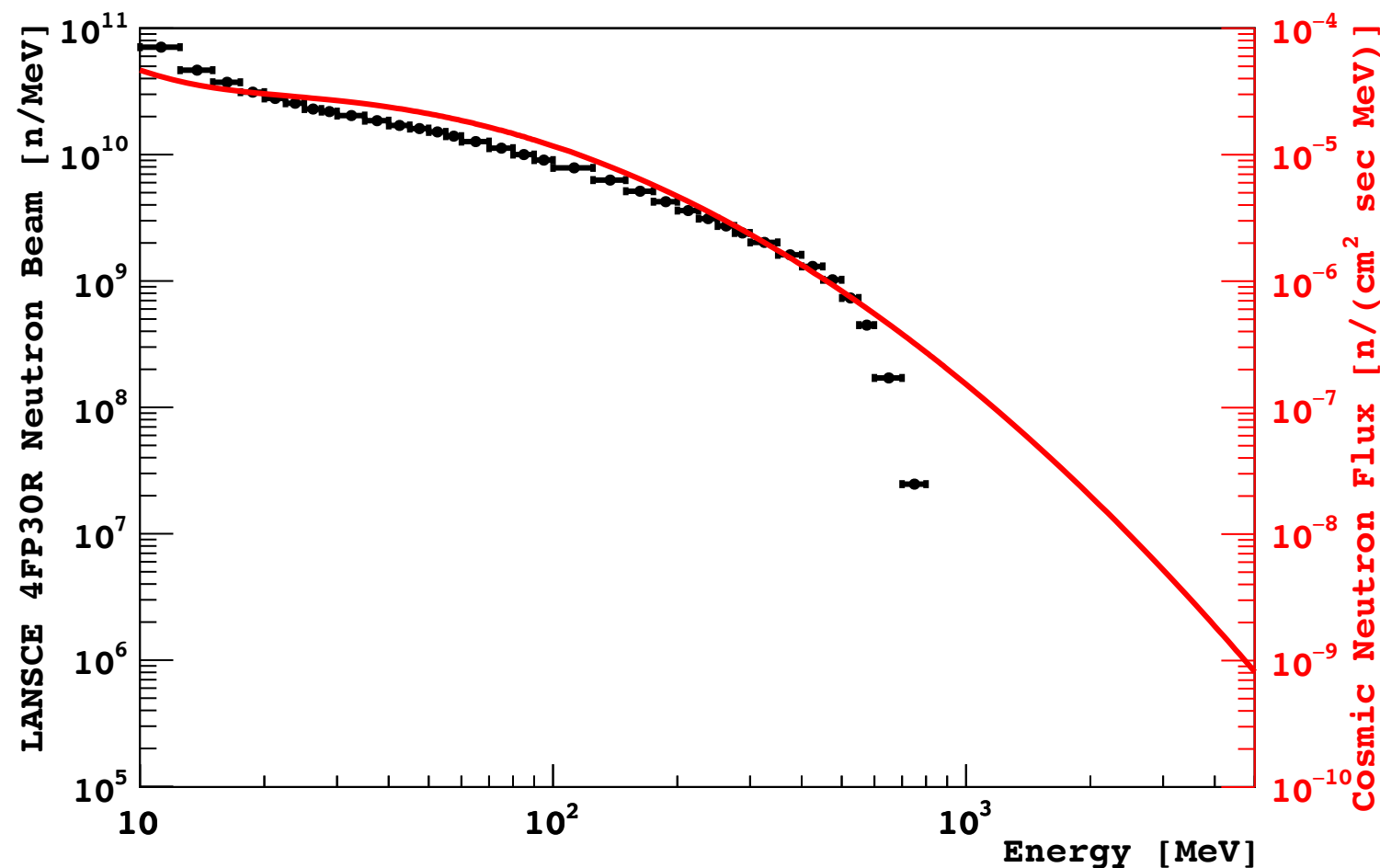
Irradiate active detector materials and use self-counting techniques to measure low-energy beta decays and x-rays

Extrapolate from measured activity to expected sea-level cosmogenic production rate



# LANSCCE ICE-HOUSE Neutron Beam

Los Alamos Neutron Science Center (LANSCCE) Weapons Neutron Research (WNR) Facility has a neutron beam (4FP30R ICE-HOUSE II) that is very similar in spectral shape to the cosmic ray spectrum



The good agreement in spectral shape between 10–500 MeV allows for low-uncertainty extrapolations to cosmic ray activation rates

The neutron flux is roughly  $5 \times 10^8$  times larger than the sea-level cosmic neutron flux

1 second on beam  
~ 16 years on the surface



# Activation Measurements for Dark Matter Experiments

$^{39}\text{Ar}$ ,  $^{37}\text{Ar}$  in Argon

Phys. Rev. C 100, 024608 (2019)  
arXiv:1902.09072



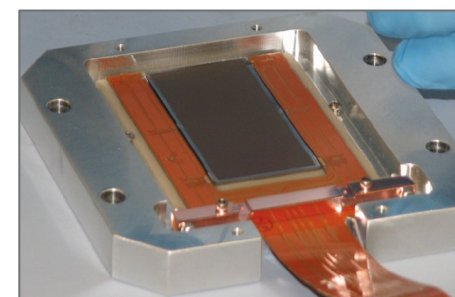
DarkSide 50



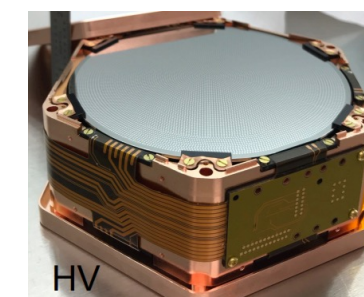
DEAP 3600

$^3\text{H}$ ,  $^7\text{Be}$ ,  $^{22}\text{Na}$  in Silicon

Phys. Rev. D 102, 102006 (2020)  
arXiv:2007.10584



DAMIC



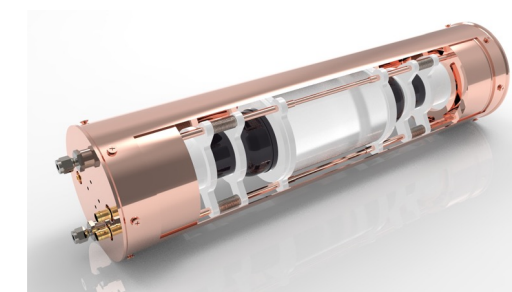
SuperCDMS

$^3\text{H}$ ,  $^{109}\text{Cd}$ ,  $^{125}\text{I}$  in Sodium Iodide

Beam Time Nov 2019  
Analysis Underway



COSINE



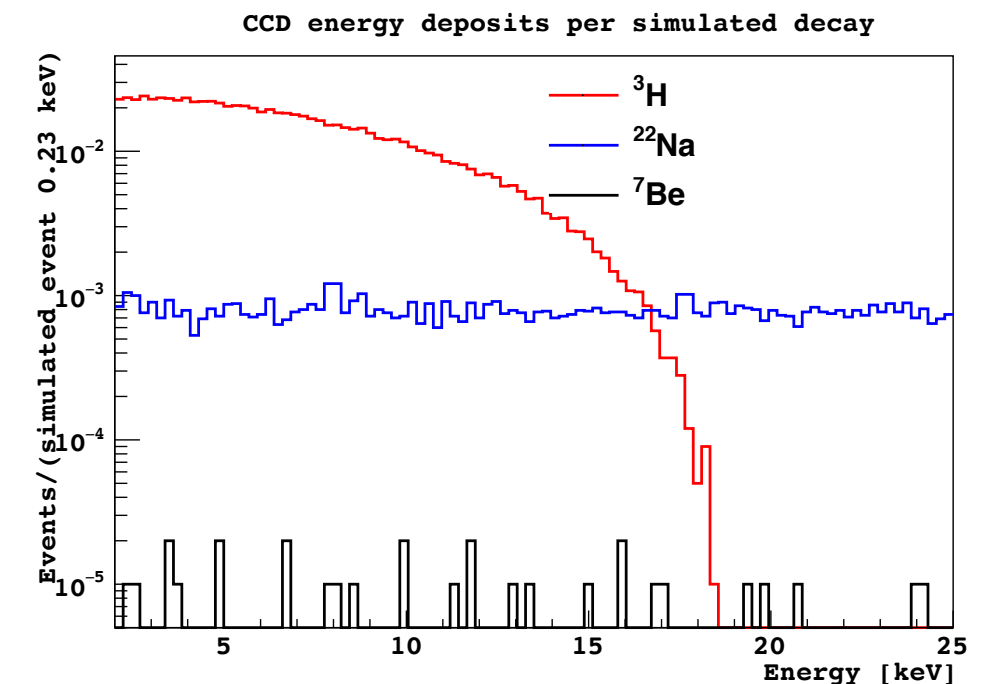
SABRE

# Cosmogenic Activation of Silicon

- Searched through Table of Isotopes for all isotopes that are lighter than Si + n/p and have a half-life of more than 30 days (also checked for radioactive daughters)
- Isotopes with half-lives greater than 100 years will not build up enough activity during the typical above-ground exposure of silicon detectors (< 10 yrs) to be a significant source of background
- $^3\text{H}$ : Low-energy pure beta-emitter, most dangerous isotope for dark matter search
- $^7\text{Be}$ : Electron-capture + 480 keV gamma, very unlikely to deposit energy in CCD
- $^{22}\text{Na}$ : Positron emitter + 1274 keV gamma, low-level background from continuous positron spectrum

*All Si activation products with half-life >30 days and any of their radioactive daughters*

Isotope	Half-Life [yrs]	Decay Mode	Q-value [keV]
$^3\text{H}$	$12.32 \pm 0.02$	$\beta^-$	$18.591 \pm 0.003$
$^7\text{Be}$	$0.1457 \pm 0.0020$	EC	$861.82 \pm 0.02$
$^{10}\text{Be}$	$(1.51 \pm 0.06) \times 10^6$	$\beta^-$	$556.0 \pm 0.6$
$^{14}\text{C}$	$5700 \pm 30$	$\beta^-$	$156.475 \pm 0.004$
$^{22}\text{Na}$	$2.6018 \pm 0.0022$	$\beta^+$	$2842.2 \pm 0.2$
$^{26}\text{Al}$	$(7.17 \pm 0.24) \times 10^5$	EC	$4004.14 \pm 6.00$

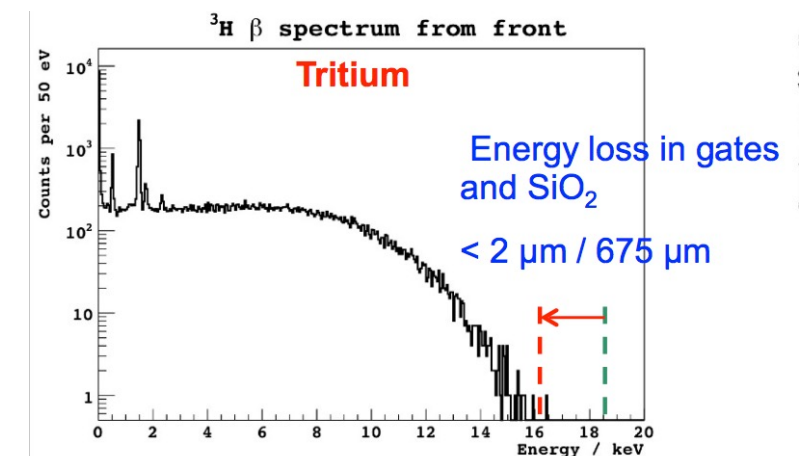
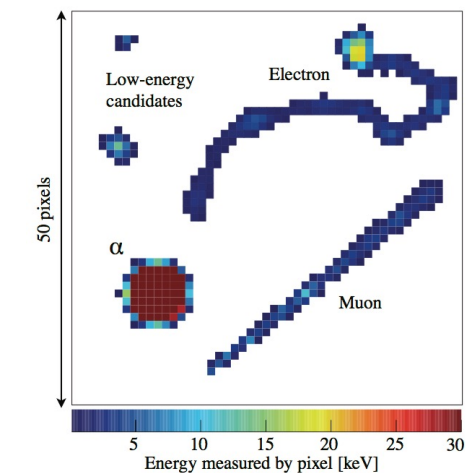
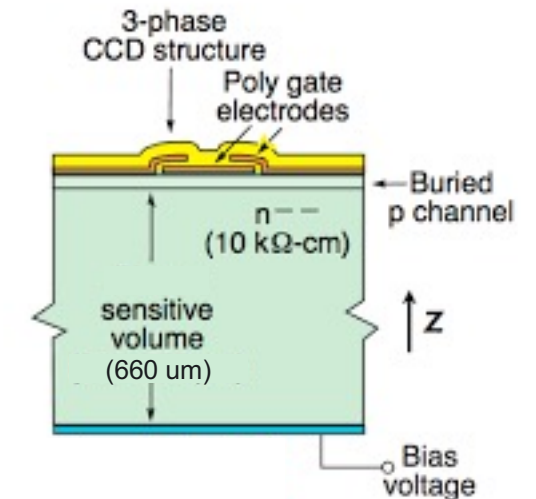




# Beam Targets

- Use **silicon wafers** to measure the gamma-ray emissions from  $^{22}\text{Na}$  and  $^7\text{Be}$  and look for any other gamma-emitters
- Use **silicon CCDs** to measure the low-energy tritium beta decays and look for any other beta decays
- CCD and readout can undergo radiation damage in beam affecting dark current and charge transfer – trade off between activation and CCD performance
- Based on previous tests with SNAP CCDs\* we aimed to keep maximum dose  $< 2 \times 10^8$  MeV/g
- We used 3 CCDs on a 1" beam with staggered exposures to ensure we had at least one CCD that survived

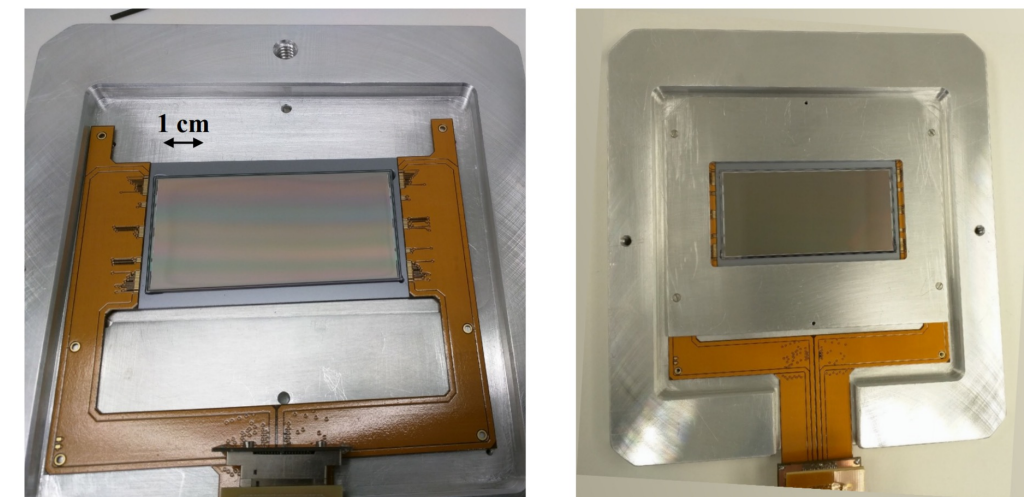
CCDs have demonstrated energy thresholds, resolution, and linearity for tritium detection



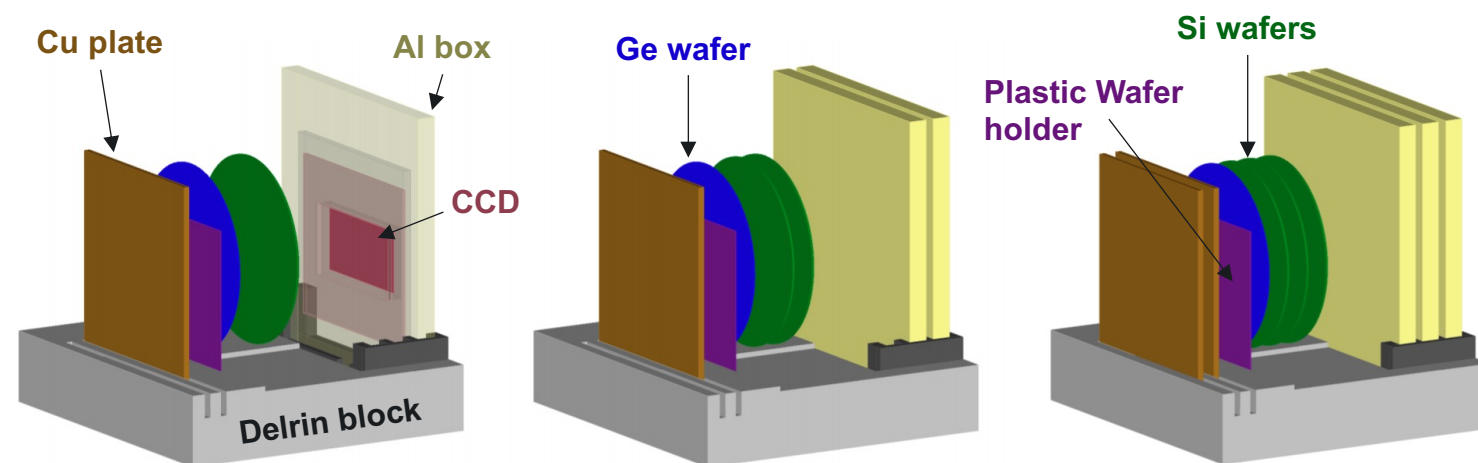
# Beam Targets

Unsure about CCD performance vs. neutron damage  
 → 3 CCDs exposed to beam for different durations

*DAMIC CCD in Protective Al Box*



*CCD kept in protective Al Box on beam line to reduce possibility of mechanical and ESD damage*

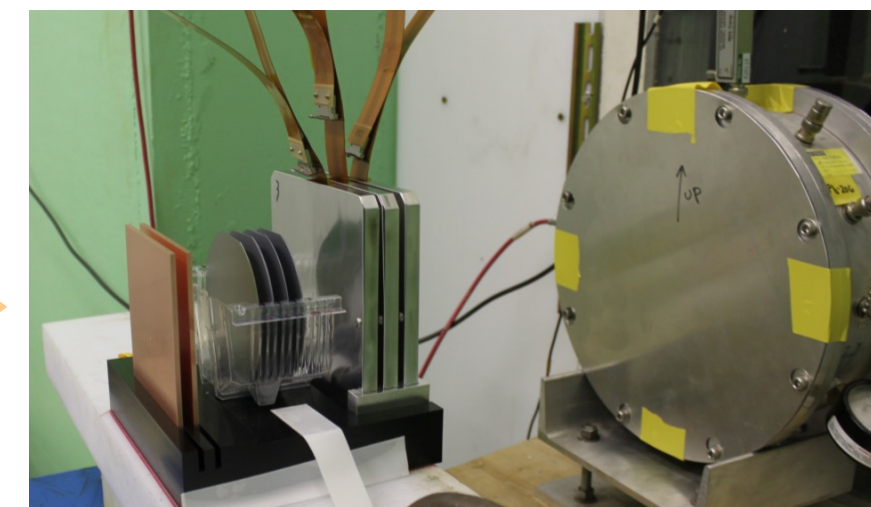


Long exposure  
samples

Long + medium  
exposure samples

Long + medium + short  
exposure samples

Target	Exposure Time [hrs]	Neutrons through target ( $> 10$ MeV)
CCD 1	109.4	$(2.39 \pm 0.18) \times 10^{12}$
Wafer 1	109.4	$(2.64 \pm 0.20) \times 10^{12}$
CCD 2	62.7	$(1.42 \pm 0.11) \times 10^{12}$
Wafer 2	62.7	$(1.56 \pm 0.12) \times 10^{12}$
CCD 3	22.8	$(5.20 \pm 0.39) \times 10^{11}$
Wafer 3	22.8	$(5.72 \pm 0.43) \times 10^{11}$

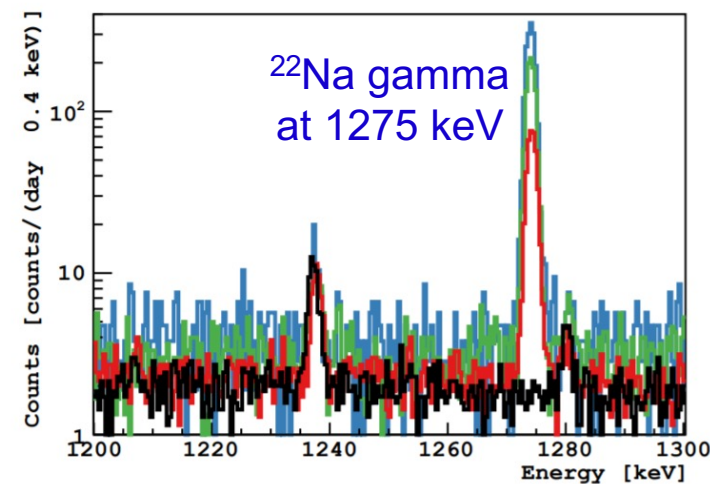
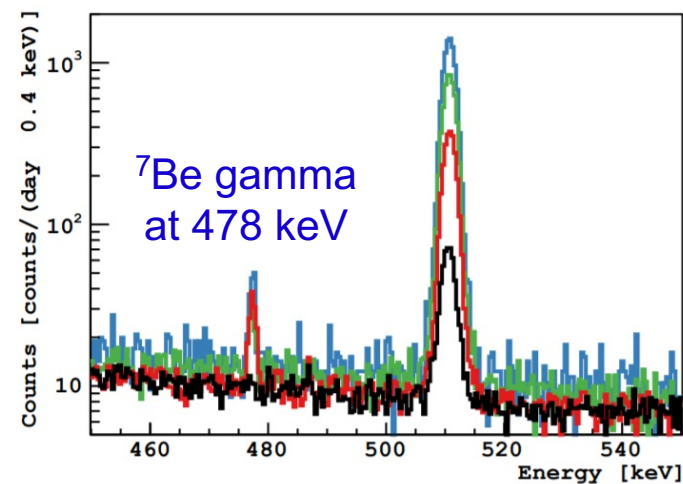
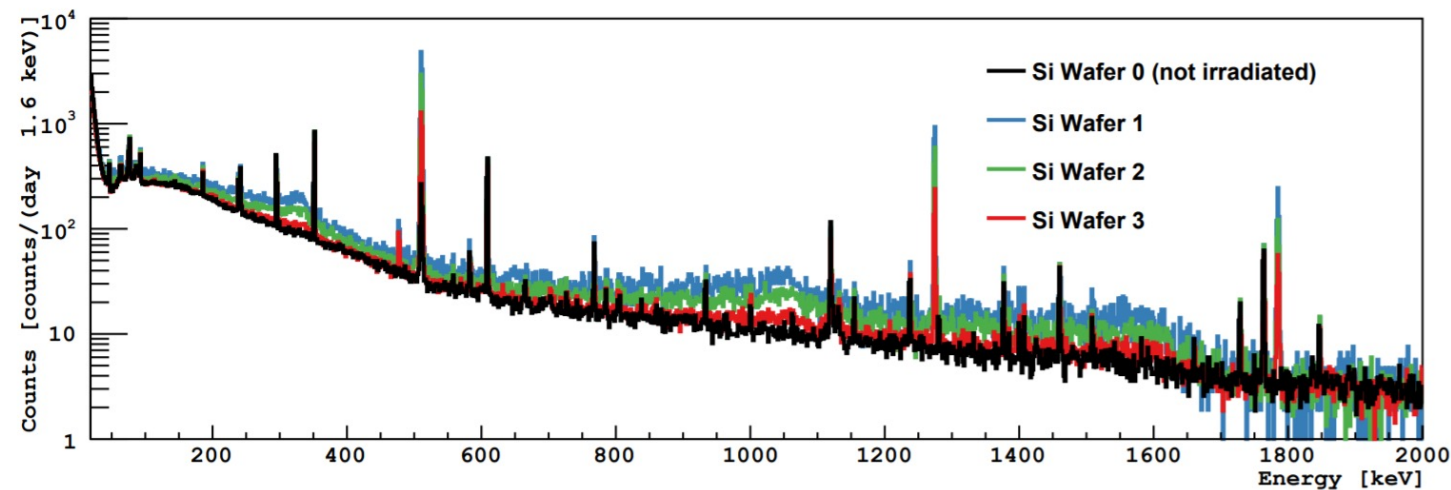


*Targets on beam line at LANSCE, LANL*



# Silicon Wafer Counting

- Each CCD was paired with two Si wafers that were exposed for the same duration on the LANSCE beam
- Wafer pairs were gamma counted at PNNL on a HPGe detector in underground lab



- $^7\text{Be}$  and  $^{22}\text{Na}$  peaks visible above background
- No other peaks observed relative to unexposed Si control wafers (black spectrum)
- Lack of a peak at 1809 keV constrains  $^{26}\text{Al}$  activation at the MDA level of HPGe detector
  - ~58x lower than  $^{22}\text{Na}$  activation rate in wafer pair #1 (i.e., the longest-exposure wafers)

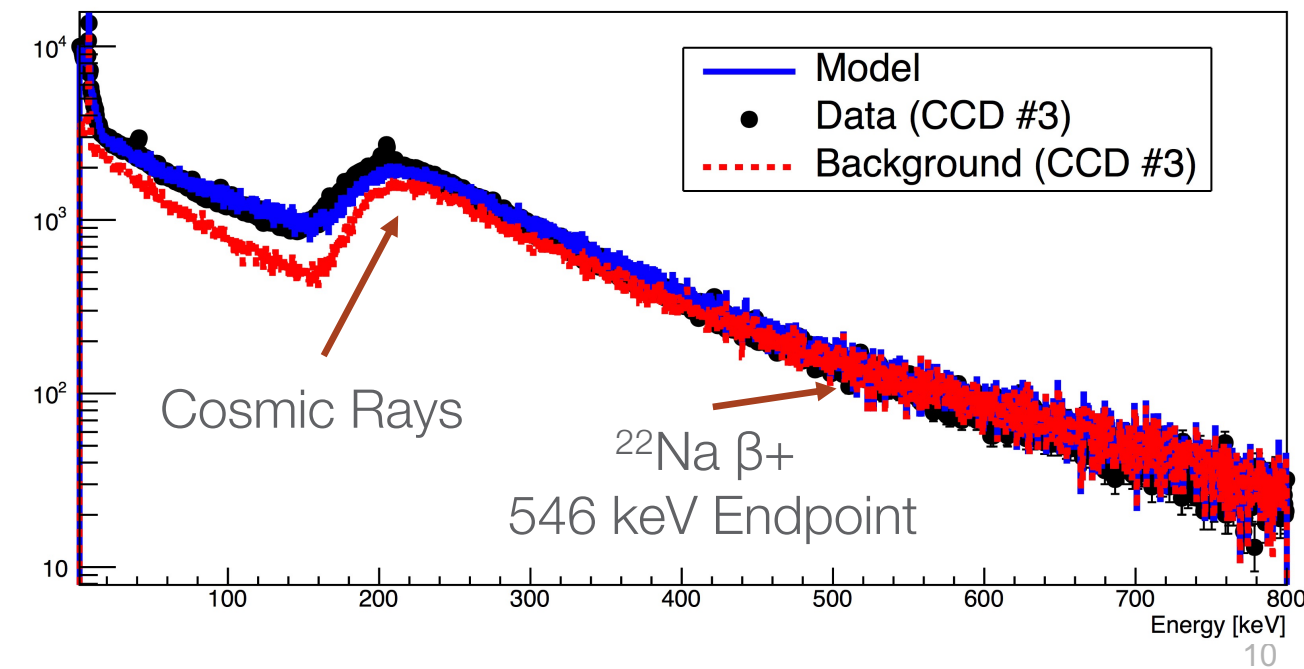
	Wafer 0	Wafer 3
Si areal density [atoms/cm <sup>2</sup> ]	-	82.342
Beam to meas. time [days]	-	7.000
Ge counting time [days]	7.000	7.000
Measured $^7\text{Be}$ activity [mBq]	<40	149 ± 12
Decay-corrected $^7\text{Be}$ activity [mBq]	-	437 ± 34
Beam-avg. $^7\text{Be}$ cross section [cm <sup>2</sup> ]	-	$(1.01 \pm 0.12) \times 10^{-27}$
Measured $^{22}\text{Na}$ activity [mBq]	<5.1	139.5 ± 6.3
Decay-corrected $^{22}\text{Na}$ activity [mBq]	-	148.2 ± 6.6
Beam-avg. $^{22}\text{Na}$ cross section [cm <sup>2</sup> ]	-	$(6.15 \pm 0.58) \times 10^{-27}$

# CCD Counting

- CCDs operated at University of Chicago surface lab
- Individually self-counted & compared to pre-exposure background levels
- All three CCDs show clear evidence of tritium betas and  $^{22}\text{Na}$  positrons
- Increased CTI and dark rate required modifications to standard DAMIC analysis
- CCD3 (least irradiated CCD) was the most straightforward to analyze and is basis of the tritium measurement



*Full energy spectrum*

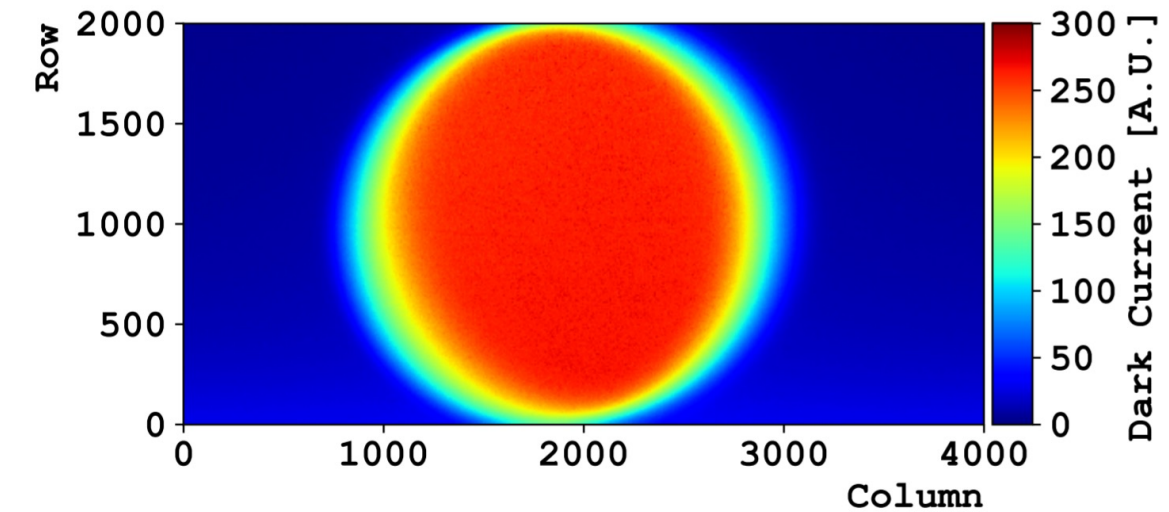




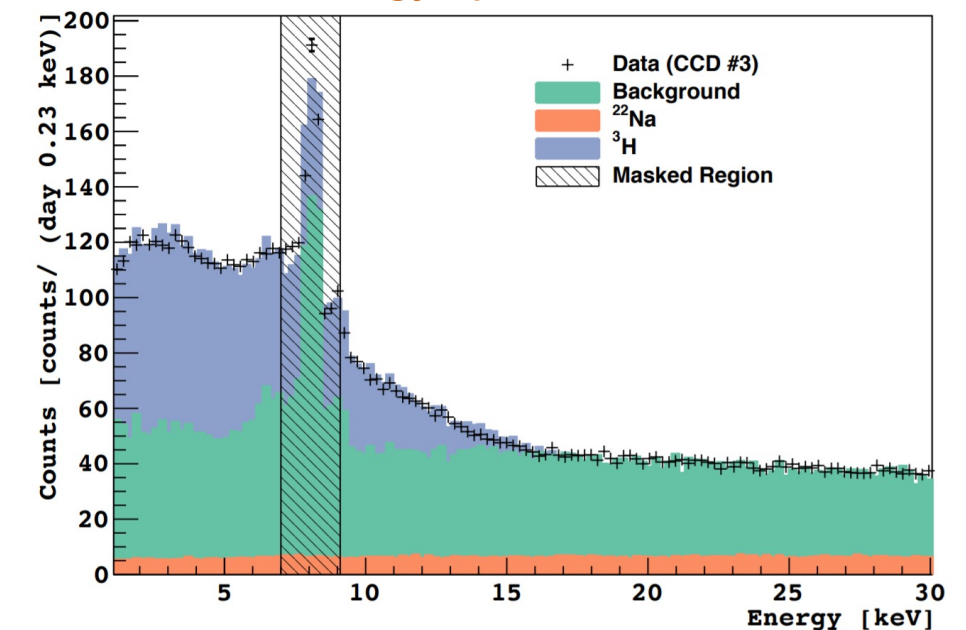
# CCD Analysis

- Simulated “blank” images were created with the same noise and dark-current profile as post-irradiation data
- Ionization from Geant4-simulated decays added to images before applying full reconstruction code and analysis
- Operation, readout, analysis all follow standard DAMIC methods
- CTI modeled with constant Poissonian kernel representing charge loss for each vertical transfer ( $\sim 9 \times 10^{-4}$ )
- Binned Poissonian likelihood fit of energy spectrum to determine contributions of  $^3\text{H}$  and  $^{22}\text{Na}$

Short-exposure CCD (#3) dark current profile



Low-energy spectrum and fit



Best Fit Parameters:

Tritium:  $45.7 \pm 0.5$  (stat)  $\pm 1.5$  (syst) mBq

$^{22}\text{Na}$ :  $126 \pm 5$  (stat)  $\pm 26$  (syst) mBq  
(expectation from wafer:  $(88.5 \pm 5.3)$  mBq)

# Cosmogenic Neutron Production Rates

## What we are trying to calculate

$$P_C = \int \Phi_C(E) \cdot \sigma(E) \cdot n \cdot dE$$

$P_C$  is the cosmogenic production rate

$\Phi_C(E)$  is the Cosmogenic neutron flux

$\sigma(E)$  is the production cross-section (UNKNOWN)

$n$  is the number of silicon targets per unit mass

## What we have measured

$$P_L = \int \Phi_L(E) \cdot \sigma(E) \cdot n \cdot dE$$

$P_L$  is the LANSCE production rate

$\Phi_L(E)$  is the LANSCE neutron flux

IF the LANSCE neutron spectral shape was the same as the cosmogenic neutron spectral shape, then we wouldn't need to care about the cross-sections

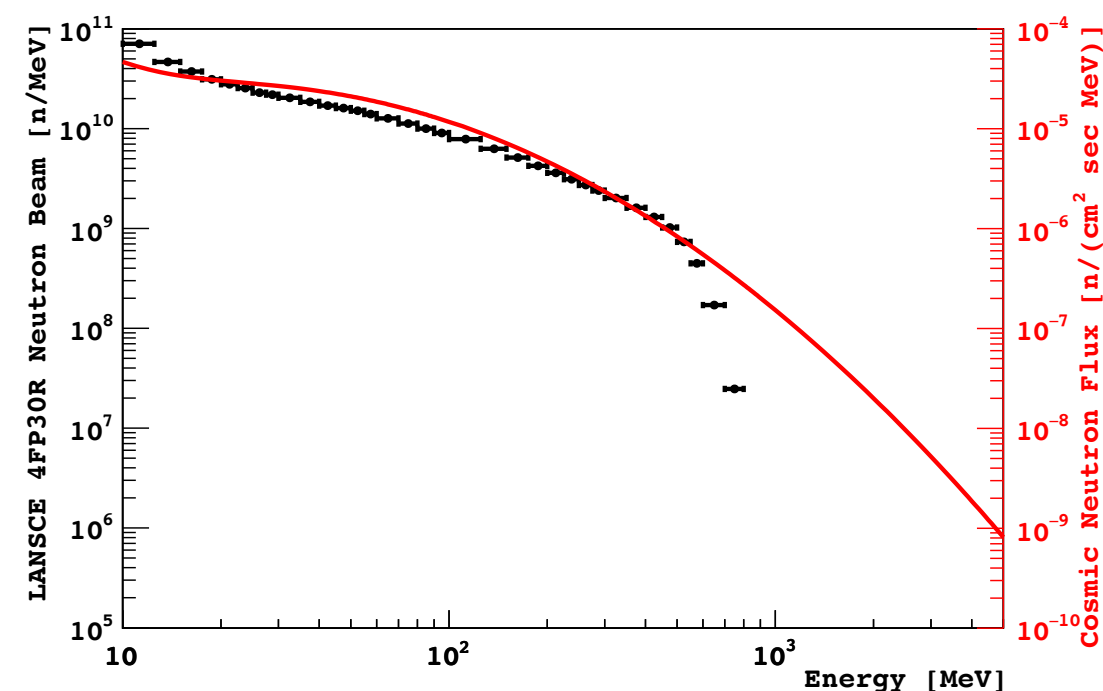
$$\text{If } \Phi_L = k \cdot \Phi_C$$

$$\text{Then } P_C = P_L / k$$

Regardless of  $\sigma(E)$

But it is not at high energies, so

**we need a cross-section model to extrapolate from  $P_L$  to  $P_C$**



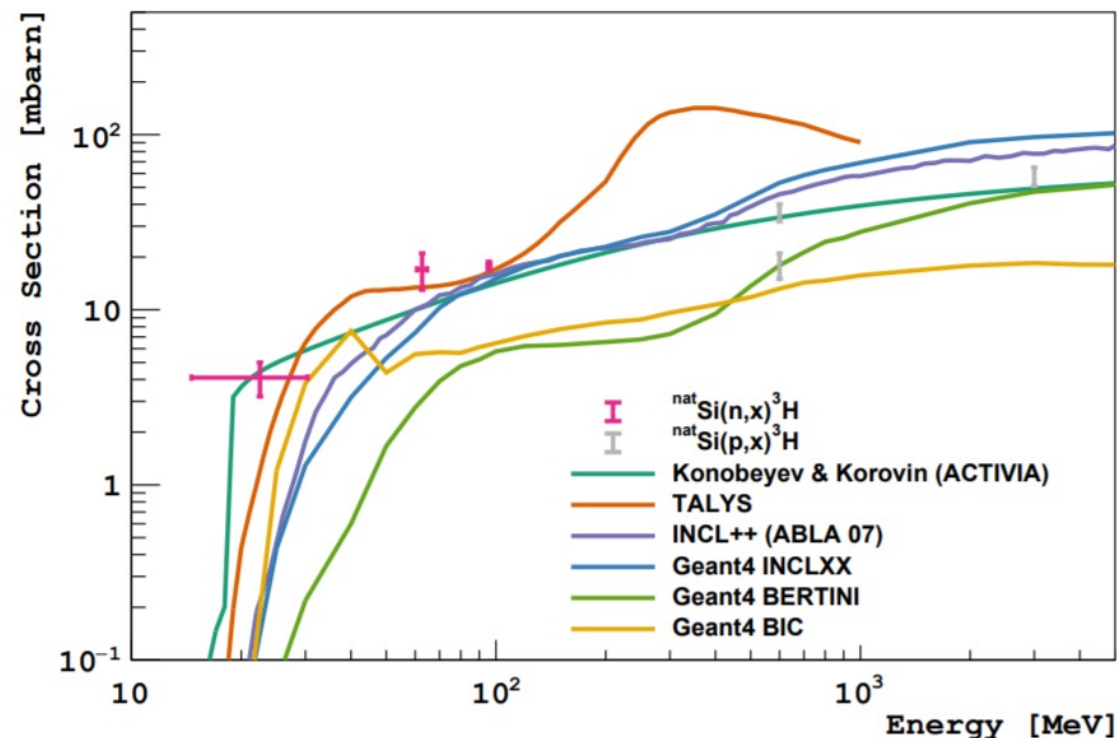


# Cross-section Dependence on Neutron Energy

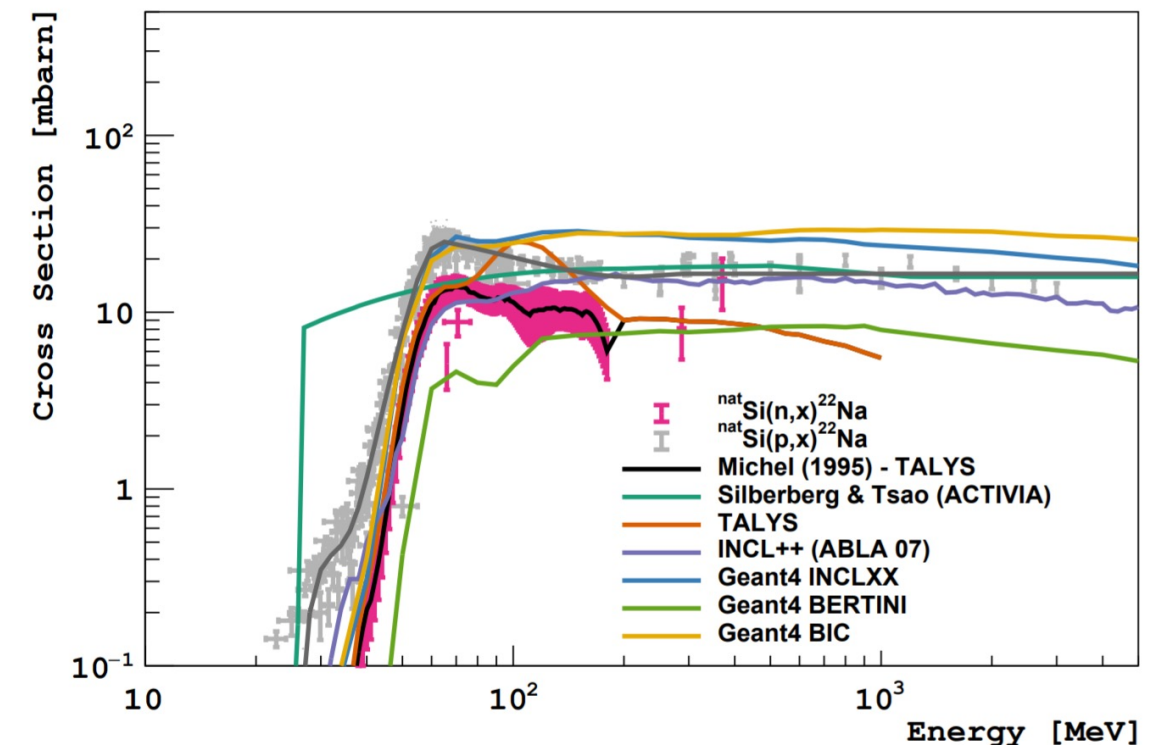
- Several models for the tritium production cross-section energy dependence  
Large difference between models ... we don't know which one is correct
- Beam measurement integrates over entire spectrum:  
(i.e., no good way to unfold energy dependence)

$$P = \frac{n_a}{\tau} \int S(E) \cdot \sigma(E) dE$$

*<sup>3</sup>H models: energy dependence varies*



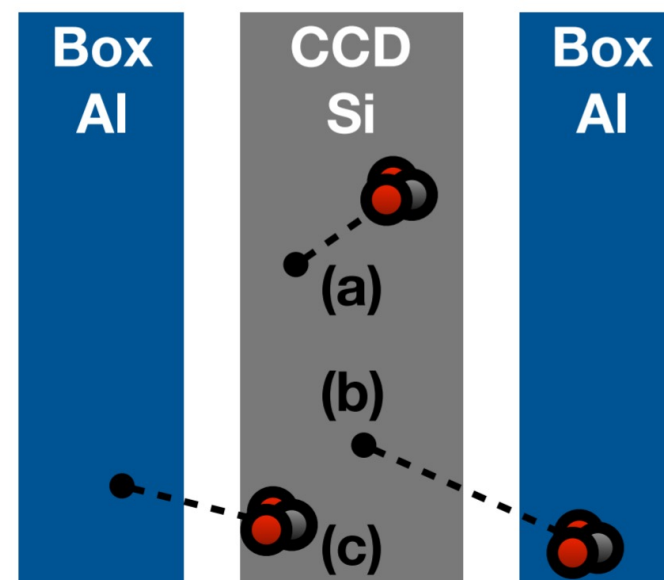
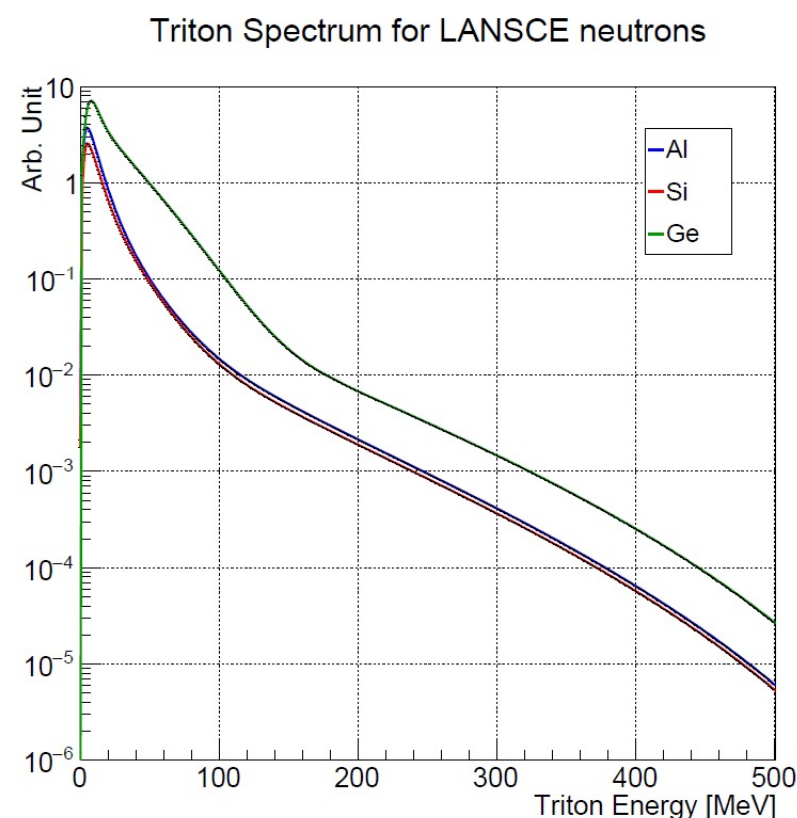
*<sup>22</sup>Na models: energy dependence very similar*



However we can use the LANSCE measurement to normalize each of the cross-section models so that they agree with the data

# Triton Ejection and Implantation

- Tritons are produced with significant kinetic energy:  
 Fraction of triton produced in Si CCD are ejected  
 Fraction produced in Al boxes are implanted into CCDs
- Fraction ejected and implanted depend on initial kinetic and angular distributions as well as geometry of beam targets – needs dedicated Geant4 simulation to evaluate.



*Geant4 INCLXX simulation of triton transfer*

Implanted	World	Ge	Si3	Si2	Si1	Box3	Box2	Box1	CCD3	CCD2	CCD1	Produced
World	20	12	4	91	50	19	42	21	6	41	1653	
Ge	1	1	0	3	2	1	17	16	9	183	5	
Si3	0	0	1	0	1	2	3	11	40	4	1	
Si2	1	2	1	2	5	3	29	111	2	5	2	
Si1	4	4	2	14	14	9	187	5	0	4	7	
Box3	2	4	8	6	22	151	1	0	0	0	1	
Box2	11	22	0	61	414	3	2	0	0	0	3	
Box1	36	1	0	693	9	0	2	0	0	0	21	
CCD3	0	1	21	1	4	10	0	0	0	0	0	
CCD2	3	59	0	9	29	0	0	0	0	0	0	
CCD1	98	0	0	47	1	0	0	0	0	0	1	
	CCD1	CCD2	CCD3	Box1	Box2	Box3	Si1	Si2	Si3	Ge	World	



# Analysis Approach

Start with multiple  
cross-section models

Predict triton activity  
ejected from CCD &  
triton activity implanted  
into CCD for each model

Calculate ratio of  
measured-to-predicted  
residual activity in CCD

Model	Pred. LANSCE <sup>3</sup> H prod. act. $P_{CCD3}$ [mBq]	Ejected Activity $E_{CCD3}$ [mBq]	Implanted Activity $I_{CCD3}$ [mBq]	Pred. LANSCE <sup>3</sup> H res. act. $R_{CCD3}$ [mBq]	Meas./Pred. <sup>3</sup> H res. act.
K&K (ACTIVIA)	40.8 ± 4.2			41.5 ± 5.4	1.10 ± 0.15
TALYS	116 ± 16	46.70 ± 0.12	53.8 ± 2.1	123 ± 17	0.370 ± 0.052
INCL++(ABLA07)	41.8 ± 4.5			42.5 ± 5.7	1.07 ± 0.15
GEANT4 BERTINI	13.0 ± 1.4	3.354 ± 0.072	3.699 ± 0.045	13.3 ± 1.5	3.43 ± 0.40
GEANT4 BIC	17.8 ± 1.7	4.995 ± 0.084	6.421 ± 0.059	19.2 ± 1.9	2.38 ± 0.25
GEANT4 INCLXX	42.3 ± 4.8	20.65 ± 0.11	16.94 ± 0.10	38.5 ± 4.4	1.19 ± 0.14

Predict total triton  
activity produced  
for each model

Predict residual  
triton activity in CCD  
for each model

*For models without ejection/implantation information, use average of other models*

# Convert to Cosmogenic Production Rate

$$P_{Ci} = \left( \frac{P_L}{\int \Phi_L(E) \cdot \sigma_i^N(E) \cdot n \cdot dE} \right) \int \Phi_C(E) \cdot \sigma_i^N(E) \cdot n \cdot dE$$

LANSCCE-measured to predicted production ratio  
(from previous slide)

Predicted cosmogenic production rate  
(uses cosmic-ray neutron spectrum instead)

This ratio normalizes each model so that it agrees  
with our measurement

Model	Meas./Pred. <sup>3</sup> H res. act.	Pred. Cosm. <sup>3</sup> H prod. rate [atoms/(kg d)]	Scaled Cosm. <sup>3</sup> H prod. rate [atoms/(kg d)]
K&K (ACTIVIA)	1.10 ± 0.15	98 ± 12	108 ± 20
TALYS	0.370 ± 0.052	259 ± 33	96 ± 18
INCL++(ABLA07)	1.07 ± 0.15	106 ± 13	114 ± 21
G4 BERTINI	3.43 ± 0.40	36.1 ± 4.5	124 ± 21
G4 BIC	2.38 ± 0.25	42.8 ± 5.4	102 ± 17
G4 INCLXX	1.19 ± 0.14	110 ± 14	130 ± 23

← This spread is assigned as a  
systematic uncertainty (±10.7%)



# Final Neutron-induced Cosmogenic Activations

**Tritium:**      **112**     $\pm 14_{exp}$      $\pm 12_{cs}$      $\pm 14_{nf}$  atoms per kg Si per day sea-level exposure

**<sup>7</sup>Be:**      **8.1**     $\pm 1.3_{exp}$      $\pm 1.1_{cs}$      $\pm 1.0_{nf}$  atoms per kg Si per day sea-level exposure

**<sup>22</sup>Na:**      **43.0**     $\pm 4.6_{exp}$      $\pm 0.4_{cs}$      $\pm 5.4_{nf}$  atoms per kg Si per day sea-level exposure

Experimental uncertainties  
(e.g., beam flux, count statistics, fits)

Neutron flux uncertainty  
(i.e. cosmic flux)

Cross-section energy dependence uncertainty  
(i.e., spread among different cs models)

# Overall Cosmogenic Production Rates

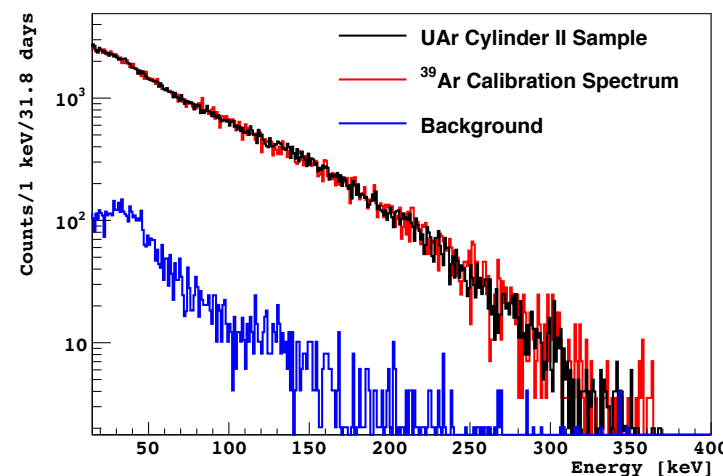
Source	$^3\text{H}$ production rate [atoms/(kg day)]	$^7\text{Be}$ production rate [atoms/(kg day)]	$^{22}\text{Na}$ production rate [atoms/(kg day)]
Neutrons	$112 \pm 24$	$8.1 \pm 1.9$	$43.0 \pm 7.1$
Protons	$10.0 \pm 4.5$	$1.14 \pm 0.14$	$3.96 \pm 0.89$
Gamma Rays	$0.73 \pm 0.51$	$0.118 \pm 0.083$	$2.2 \pm 1.5$
Muon Capture	$1.57 \pm 0.92$	$0.09 \pm 0.09$	$0.48 \pm 0.11$
Total	$124 \pm 24$	$9.4 \pm 2.0$	$49.6 \pm 7.3$

**Sea-level  $^3\text{H}$  production rate of 124 atoms/kg/day  
corresponds to a background rate of  
~ 0.002 dru/day in the 0-5 keV energy range.**

# Summary

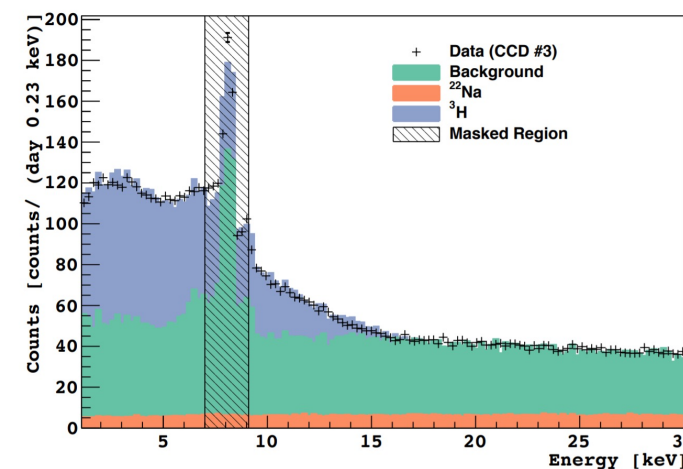
- Activation rates of critical long-lived cosmogenic isotopes are not well-known due to low production rates and low energy of decays
- We have used a method of irradiating active detector material in a high energy neutron beam and self-counting the activation products to estimate the production rates
- By irradiating silicon CCDs and wafers, we have made the first measurement of the cosmogenic production rate of  $^3\text{H}$ ,  $^7\text{Be}$ , and  $^{22}\text{Na}$  in silicon (Phys. Rev. D 102, 102006 (2020))

## Argon



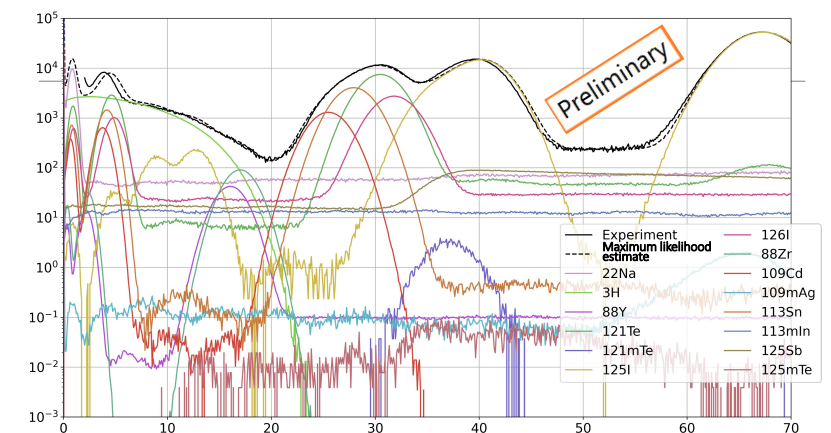
$^{39}\text{Ar}$ :  $1048 \pm 133$  atoms/kg/day  
 $^{37}\text{Ar}$ :  $92 \pm 13$  atoms/kg(AAr)/day  
 $56.7 \pm 7.5$  atoms/kg(UAr)/day

## Silicon



$^3\text{H}$ :  $124 \pm 24$  atoms/kg/day  
 $^7\text{Be}$ :  $9.4 \pm 2.0$  atoms/kg/day  
 $^{22}\text{Na}$ :  $49.6 \pm 7.3$  atoms/kg/day

## Sodium Iodide



Analysis in Progress



# Thank You

PHYSICAL REVIEW D **102**, 102006 (2020)

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## Cosmogenic activation of silicon

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