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Virtual Summer Meeting for Young Researchers

3D Printed Scintillator Metamaterials

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ETI

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NNSA

National Nuclear Security Administration

Introduction: Scintillator Metamaterials

Scintillators, materials that emit light when hit by radiation, are a backbone technology for radiation detection.

Most scintillators detect the energy of radiation-induced recoils, but not radioactivity location or emitted spectrum.

We are developing scintillator metamaterials, combinations of simple scintillators with emergent properties.

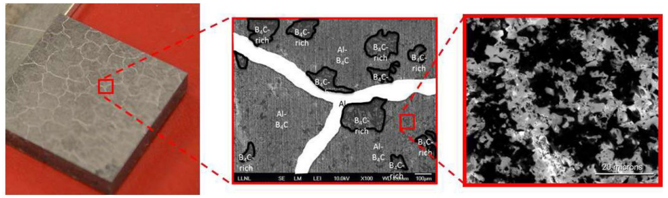
Scintillating materials can be additively manufactured (AM), though AM scintillators are an emerging technology

AM enables “metamaterials” that combine multiple materials in internal structures that produce new properties.

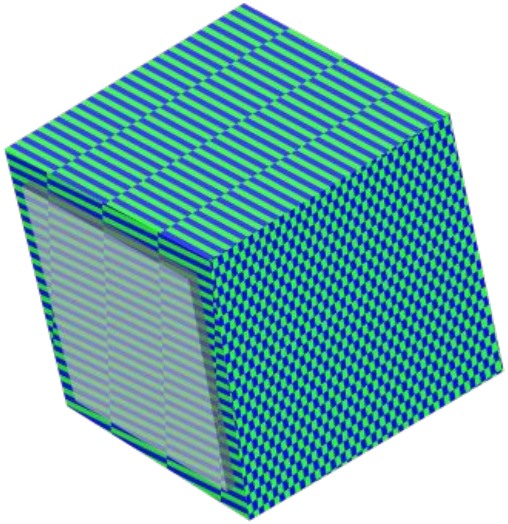
Mixed material scintillator systems (MMSS) are radiation detectors that use scintillator metamaterials to enable new capabilities.

AM rocket engine

Metal/ceramic metamaterial for armor



AM isn’t just for making unusual external shapes like rocket engines; it also enables control over internal structure of metamaterials



Controlling dye placement in scintillators creates scintillator metamaterials

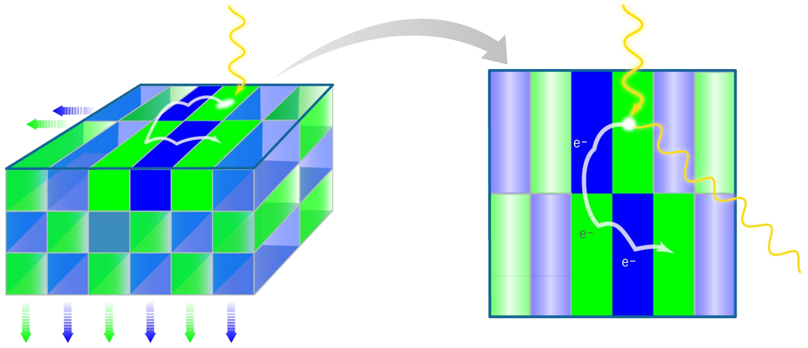
The power of scintillator metamaterials

Scintillator metamaterials can encode properties of the radiation detection event using the dye color.

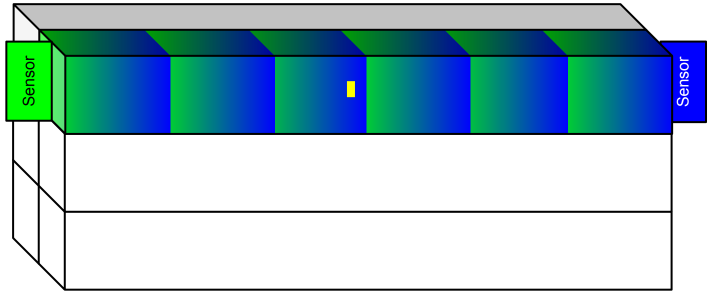
Many of the capabilities of a tracking detector or highly segmented detector.

Low cost: only two sensors per segment, and only one or a few segments needed

Using micro-scale zones, we can sense the track left by the recoil



A macroscopic gradient can measure the location of the recoil



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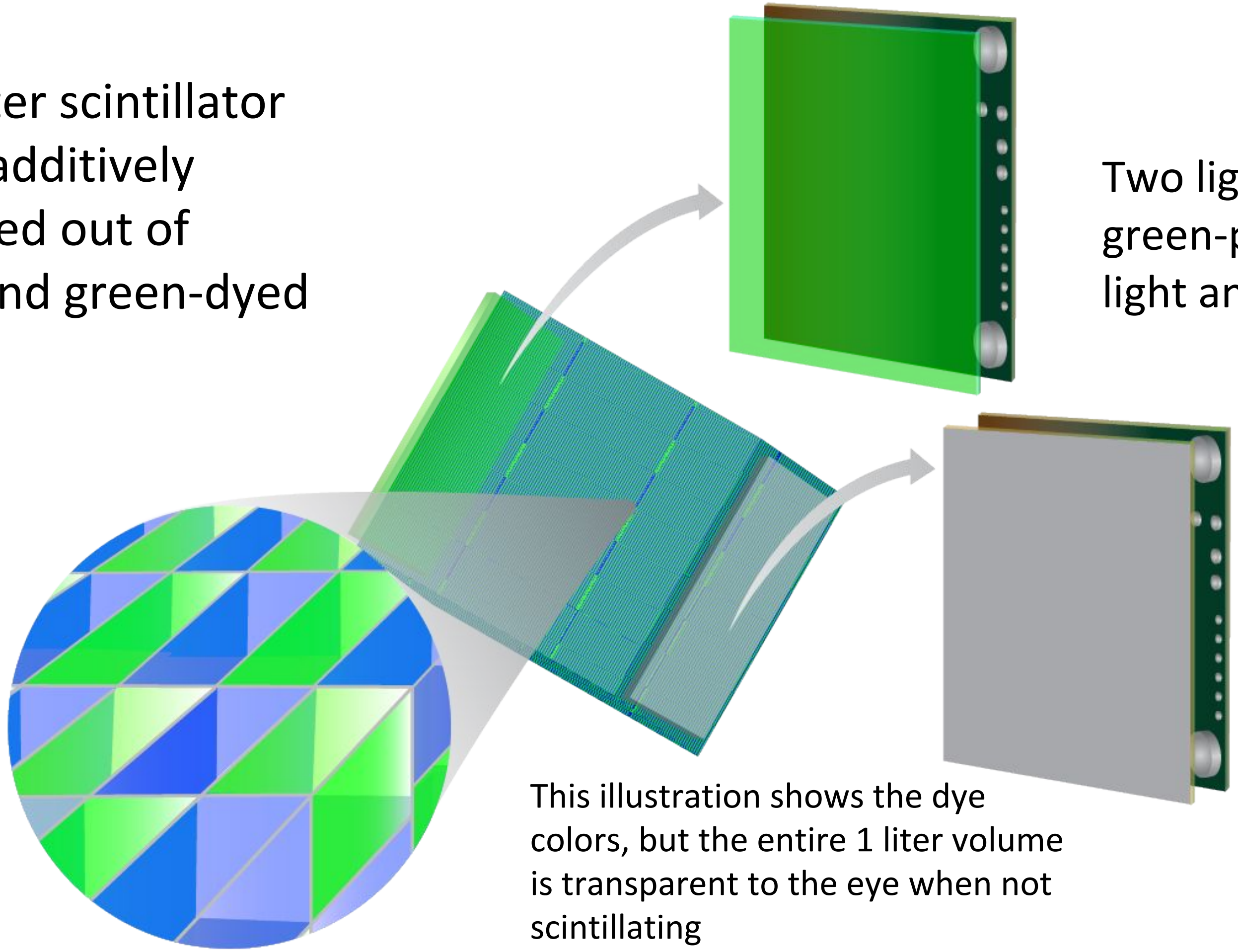
Example metamaterial design

A single 1 liter scintillator segment is additively manufactured out of blue-dyed and green-dyed scintillator.



Two light sensors, one with a green-pass filter, can measure total light and blue/green ratio.

Additive manufacturing allows for differently-dyed zones as small as 50 microns with DIW, or smaller with PµSL

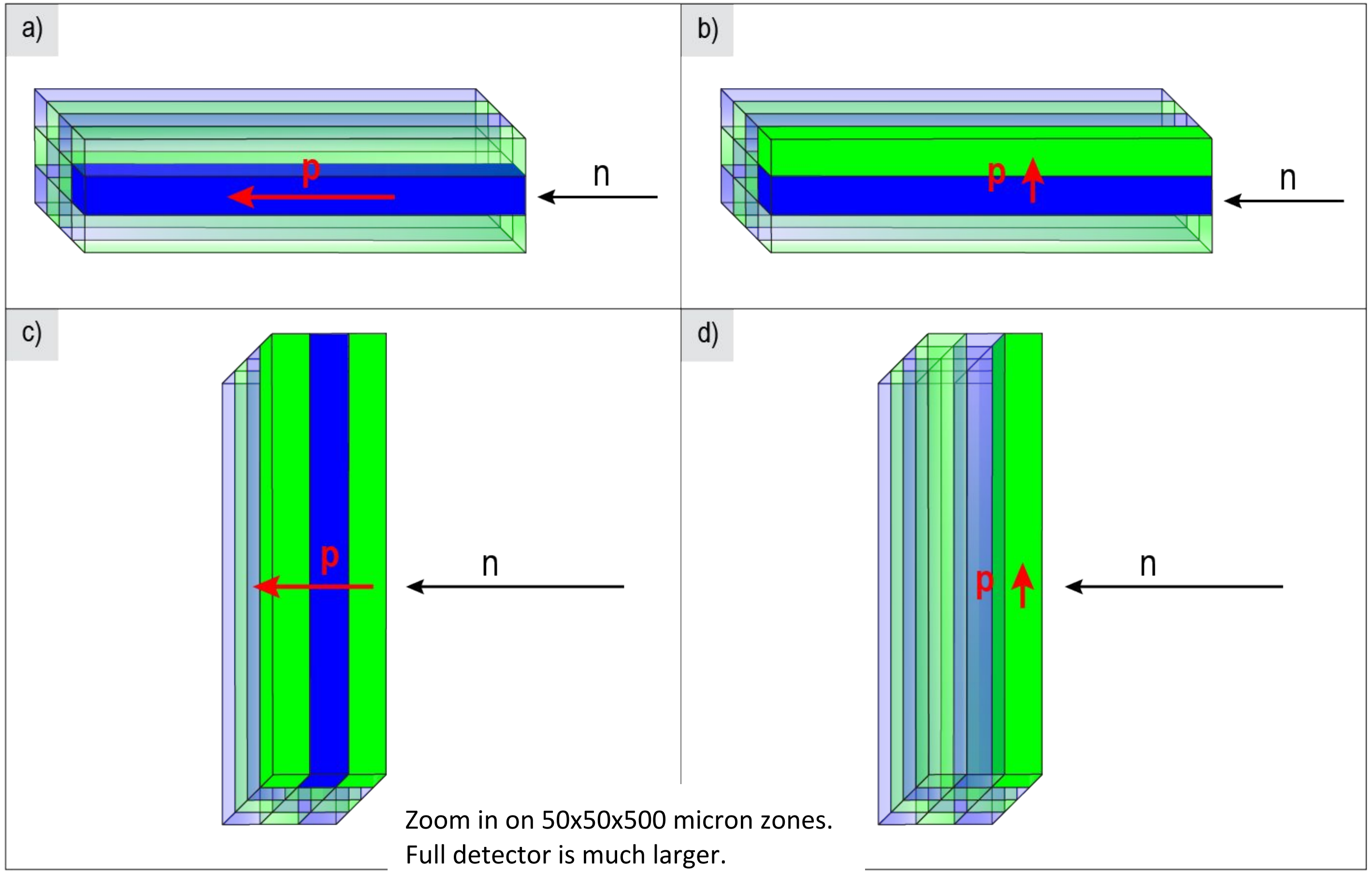
This illustration shows the dye colors, but the entire 1 liter volume is transparent to the eye when not scintillating

Neutron detection with anisotropic metamaterial

A full-energy scatter “down the grain” produces purely blue (or green) light

A full-energy scatter “across the grain” produces a mixture of green and blue light.

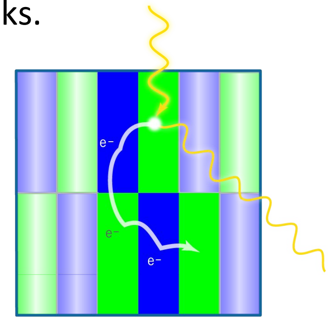
The ratio of light colors is correlated with the angle between the proton recoil and the grain.



Zoom in on 50x50x500 micron zones.
Full detector is much larger.

Large-angle scatters produce less light and short tracks that may or may not produce both colors. These scatters are generally uninformative.

Gamma backgrounds also produce both colors, due to long crooked tracks.



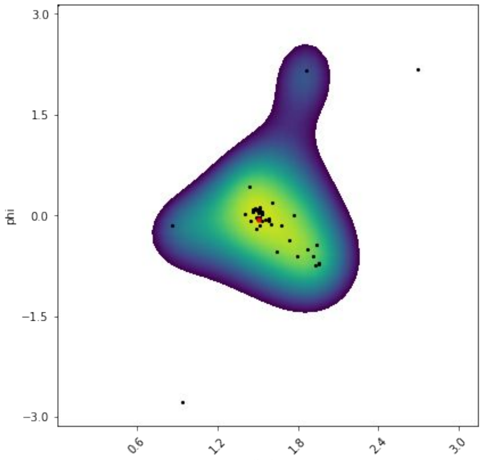
Neutron source location simulation results

Produce a large simulation set & cut away multiple scatters (opposite of scatter camera approach)

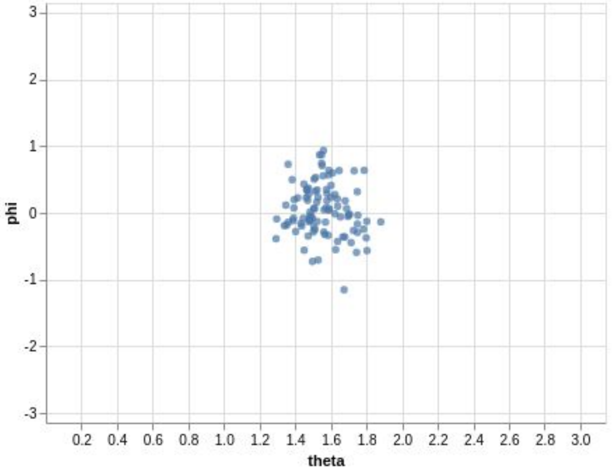
Using simulation results, we filled a 5-dimensional kernel density estimator (KDE) with:

Observables	observed photons	% in green-filtered sensor	module number
Source direction	θ	φ	

Then, we assemble a test ensemble of 50 (simulated) neutrons. We keep their observables, but try different source directions to maximize the KDE likelihood.



Gaussian process optimizer allows for efficient search for maximum likelihood



Trying several ensembles builds up a distribution of maximum likelihood locations

Neutron source location simulation results

Scenario: 10^5 neutrons/s Cf-252 at 10 m from the detector.

Efficiency after cuts: 12.6% (high, due to only requiring single scatters)

In this scenario, 1.64 neutrons/s in detector passing cuts.

N, ensemble Size	Measurement time	Angular resolution (deg)
10	6.1 s	25
50	31 s	14
250	2.5 min	8.3
1000	10 min	5.9

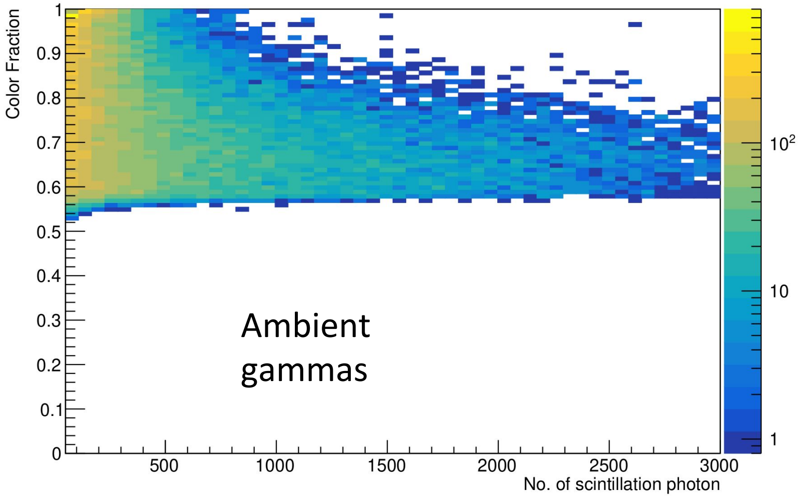
This method uses a known source spectrum. Additional work needed to make robust against unknown source spectrum.

Neutron/gamma discrimination results: Particle ID (PID)

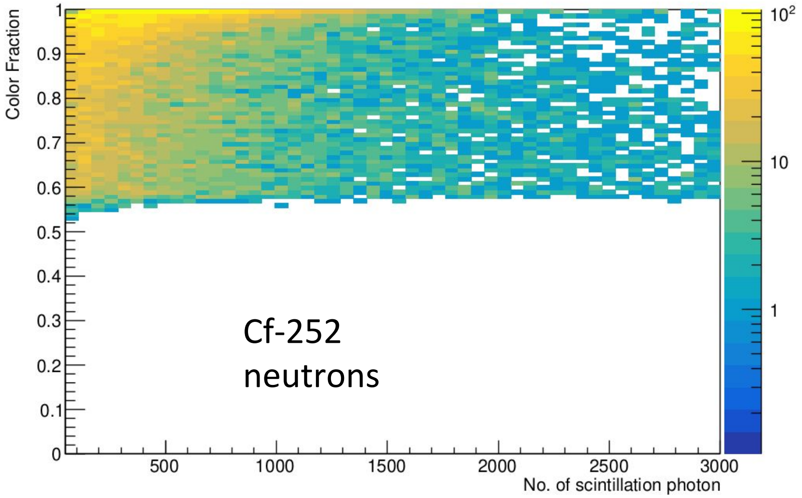
Color fraction = max(green,blue)/(green+blue)

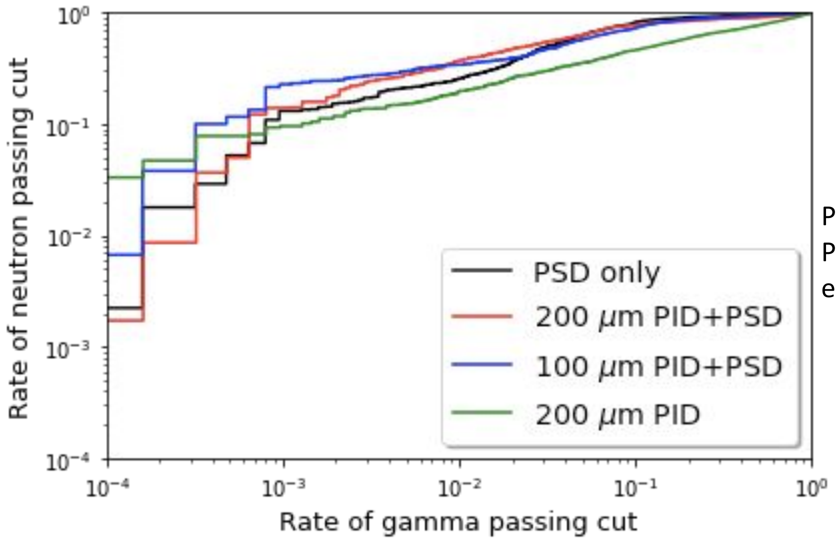
Small bias due to overlap between green and blue spectra

Gammas produce long tracks with a color fraction close to 50%, except at low energy where tracks are too short to escape zones.



Neutrons produce short tracks that stay in one zone, so have color fraction = 1 even at higher energy (except for a few which start close to a zone boundary)





PSD taken from PROSPECT experiment

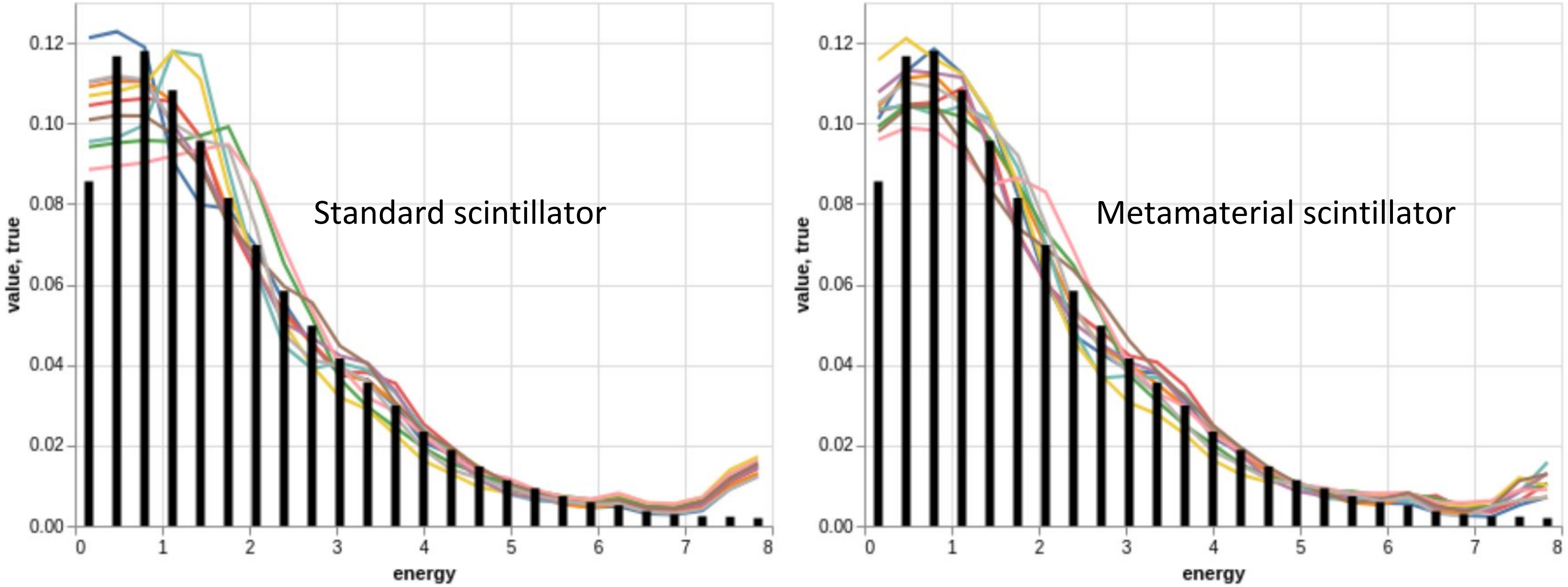
Metamaterial discrimination (PID) by itself performs worse than as-deployed PSD (and even worse than cutting-edge PSD), but using a PSD-capable metamaterial works better than either approach alone.

PSD alone: 25% neutron efficiency @ 99% gamma rejection

PSD+PID: **37**% efficiency @ 99% rejection

Spectroscopy results

Anisotropic metamaterial is sensitive to proton recoil angle, so can disambiguate neutron unfolding

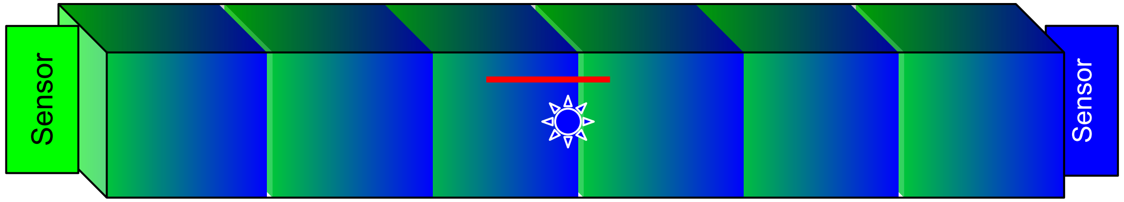


Unfolding with PyUnfold. Black: true spectrum. Colored lines: unfolded spectrum using 2000 neutrons.

SAM metric for unfolding accuracy:

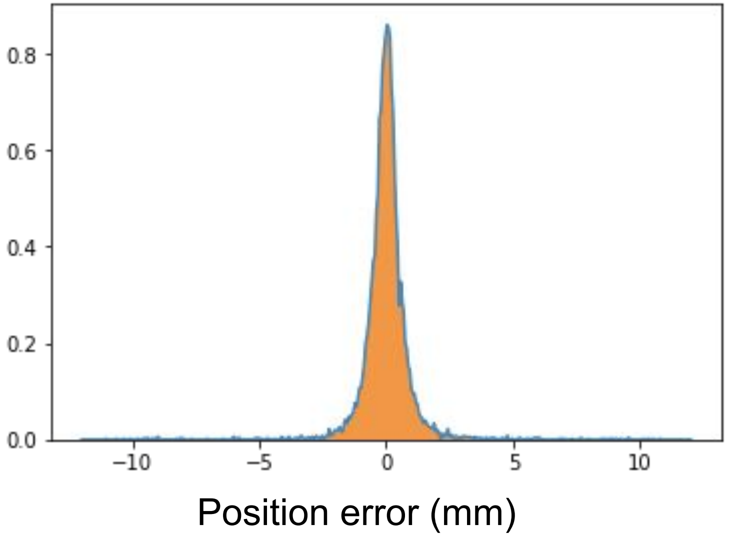
Conventional: 7.7 Metamaterial: 6.7

Hit position results (preliminary)



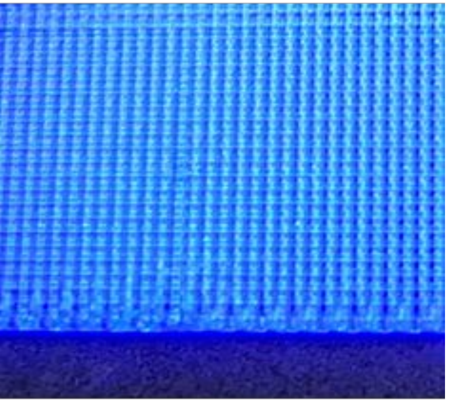
Detectors can locate the radiation interaction point along the axis of a bar of scintillator by comparing the timing and amplitude of signals sensed at both ends. This has ~1 cm precision.

Using a periodic gradient to augment this conventional approach, we can narrow the precision down to 0.8 mm.

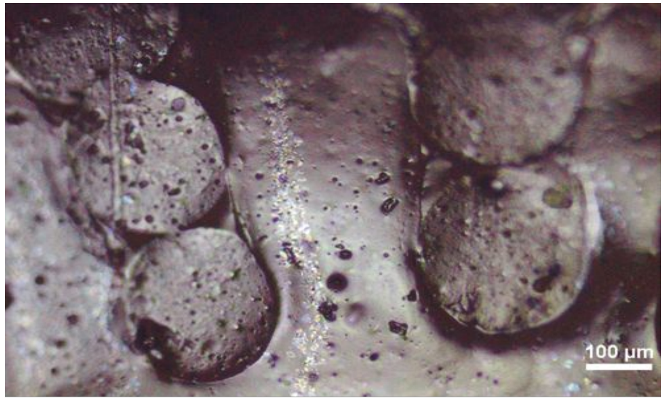


Prototyping

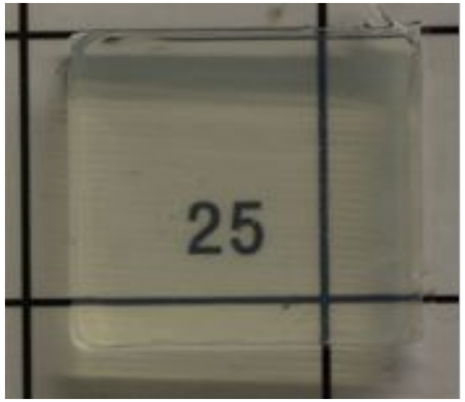
Invented polysiloxane scintillator feedstock for direct ink write:



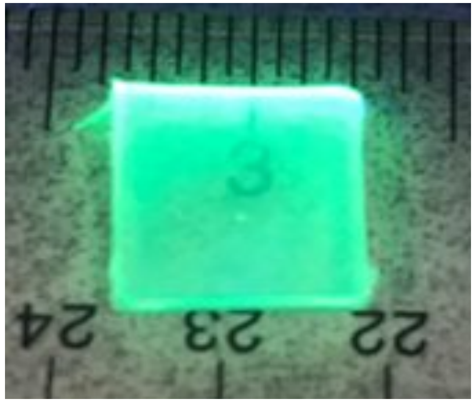
Fine structures: currently printing 250 microns, going down to 50 only requires more filtering



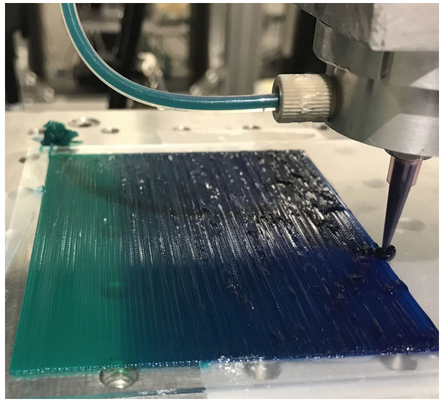
Heterogeneous structures possible via co-printing or back-filling



Optical clarity requires careful index matching



Large Stokes shift green dye prevents cross talk with blue dye



Gradient printing possible using in-nozzle mixing

Watch a video:
https://www-gs.llnl.gov/nuclear-threat-reduction/nuclear-detection-countermeasures/3D-printed-scintillator

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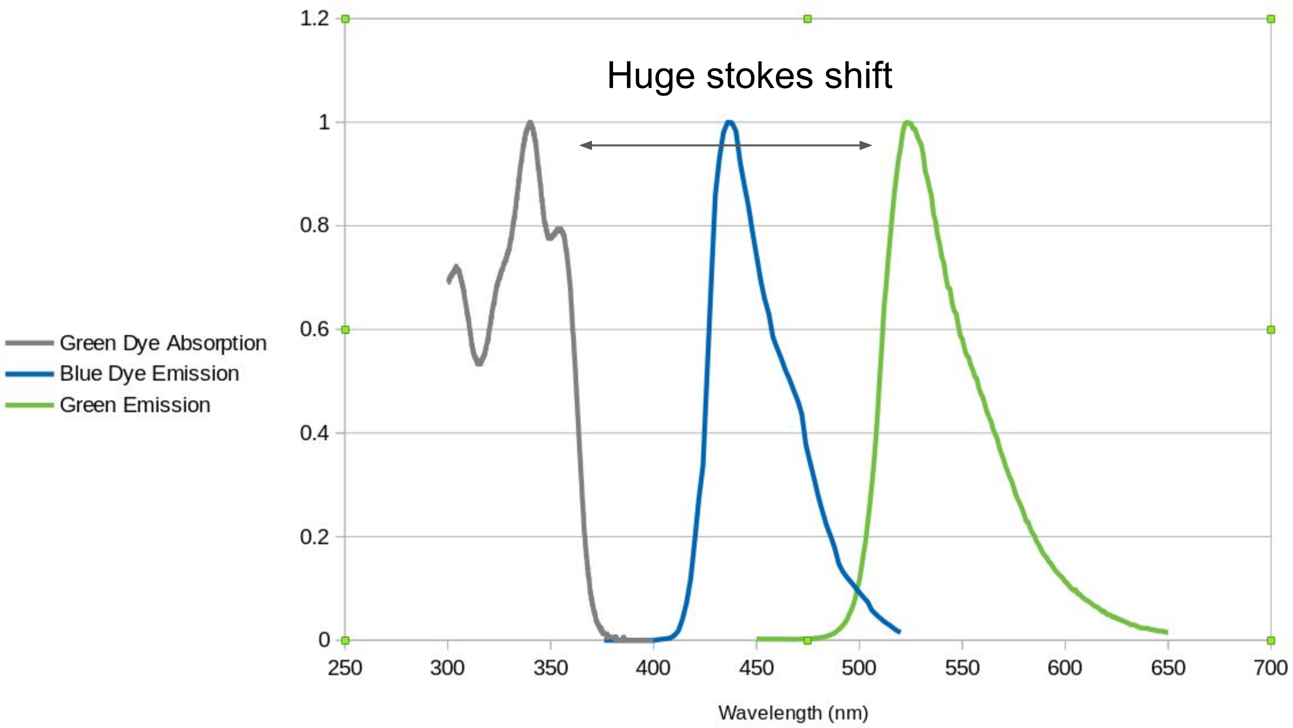
Importance of dye choice

Large stokes shift green dye enables two-color metamaterial without crosstalk.

Most green dyes would absorb blue light and reemit green, so any blue signals would be lost.



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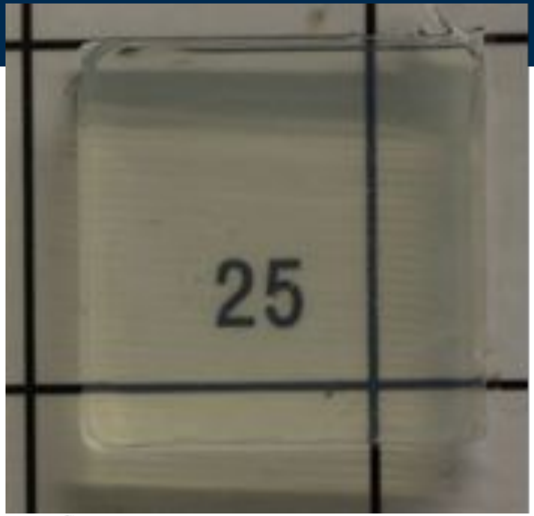
Prototyping

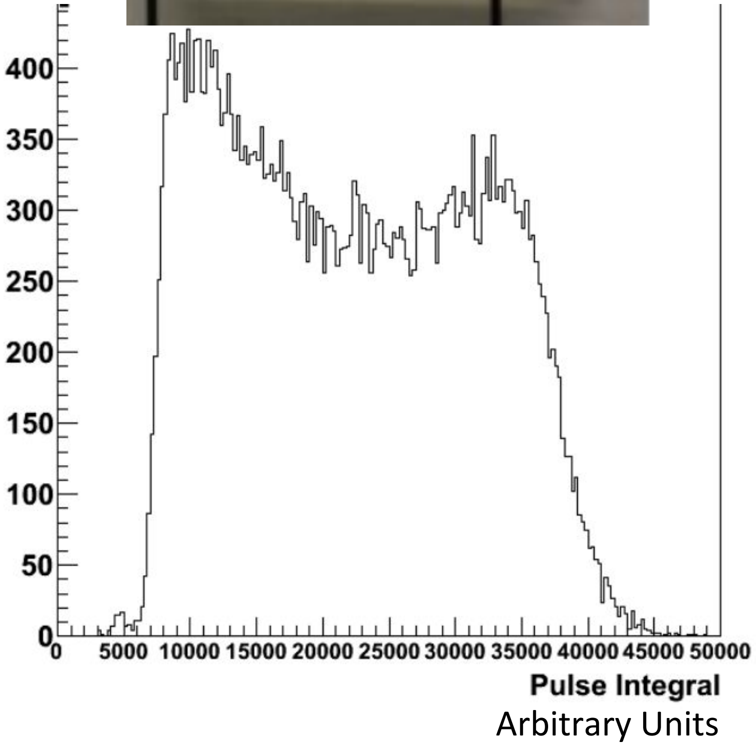


Light output measurement at 30% of EJ-200
(after correcting for green sensitivity of sensor)

0.5 g piece (scale up & optimization postponed by Covid-19)

With optimization of feedstock and technique, expect ~1 kg with at least 30% EJ-200 light output within a year.

Several other approaches possible if polysiloxane hits limitations



Conclusions

- Scintillator metamaterials encode radiation properties using additively manufactured internal structure.
- Neutron source location and spectroscopy capabilities with an anisotropic structure
- Small-scale successful printed prototypes with light output 30% of EJ-200

- Future:
  - Integrated detector prototype with kilogram of metamaterial
  - Chance to be first directional neutron detector with widespread deployment
  - Deploy for safeguards, treaty verification, antiterrorism, emergency response...

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Backup

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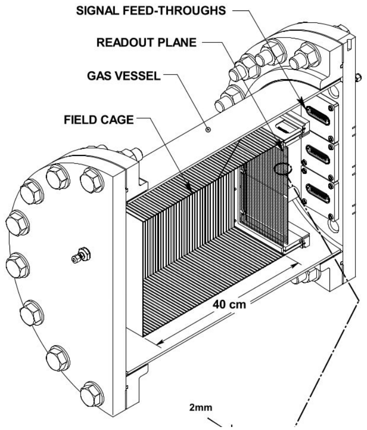
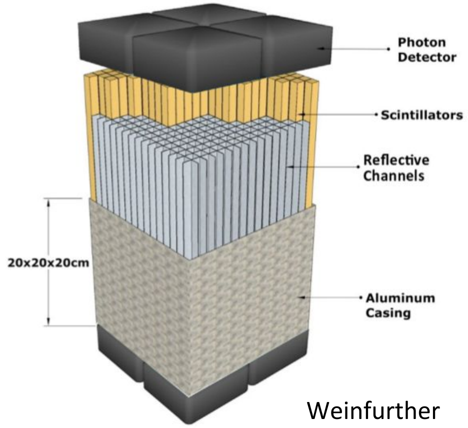
The impact of scintillator metamaterials

Tracking detectors, from ATLAS/CMS at the LHC to smaller-scale gas detectors, offer the most detailed view of radiation tracks, but have low efficiency--lots of radiation is missed, but some is detected with great detail



Bowden et al., 2010

Compact neutron scatter cameras can't see tracks, but can see multiple scatters. Powerful capabilities based on kinematic reconstruction, but high cost and medium efficiency.



Weinfurther et al., 2018

Scintillator metamaterials can emulate certain features of a tracking detector's detailed view of the track, but have high efficiency (solid, only require single scatters) and low cost (16 channels for 8 kg)

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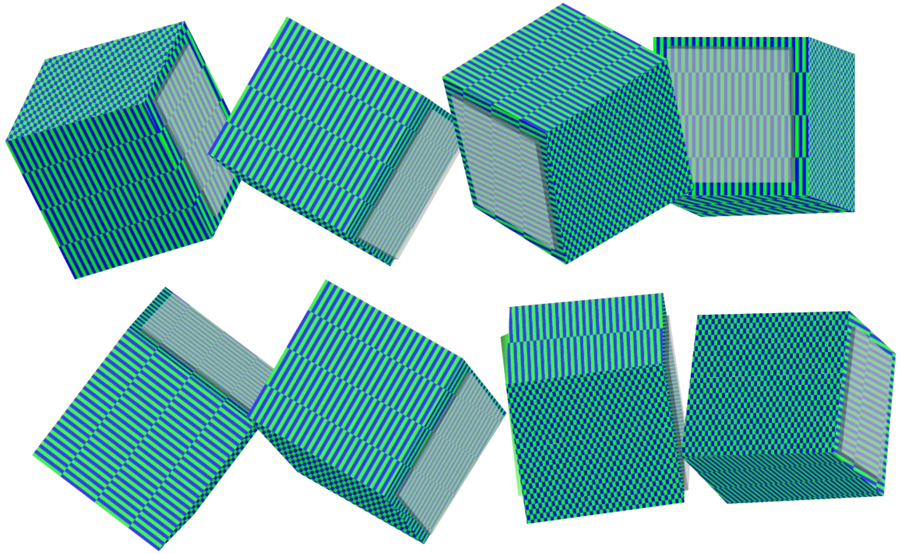
Neutron detection anisotropic array

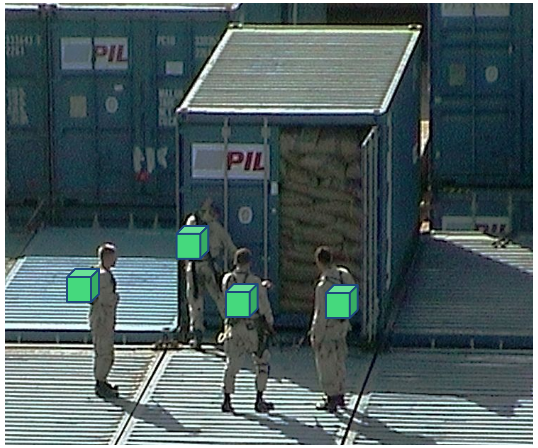
One module (1 kg) can measure the angle between recoil protons and one grain direction.

An array of ~8 modules with different grain directions can be used to locate sources in 4π

These modules can be located in a single container or in multiple containers. e.g.:

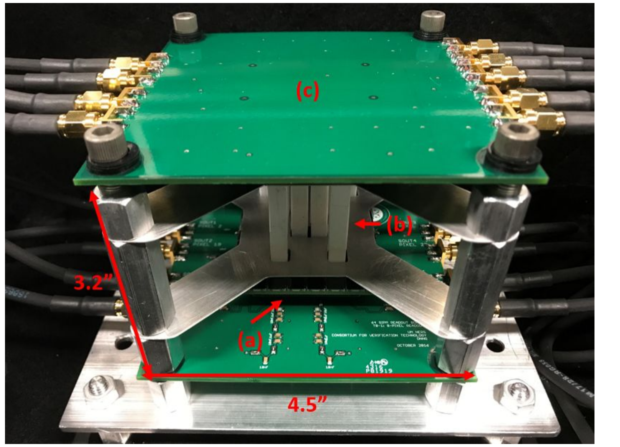
- One module in each backpack of a search team
- Modules surrounding a portal





Comparison to compact neutron scatter cameras

Compact neutron scatter cameras: more power per neutron, but lower efficiency produces similar resolution at a given measurement time:



1.46% efficiency in an 8 kg detector

K. Weinfurther, J. Mattingly, E. Brubaker, and J. Steele, “Model-based design evaluation of a compact, high-efficiency neutron scatter camera,” *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, vol. 883, pp. 115–135, Mar. 2018, doi: 10.1016/j.nima.2017.11.025.

13.2 & 14.7 degree resolution (azimuth & elevation) with 5 neutrons

W. M. Steinberger, M. L. Ruch, N. P. Giha, A. Di Fulvio, S. D. Clarke, and S. A. Pozzi, “Low-Statistics Imaging of Weapons-Grade Plutonium using a Handheld Neutron Scatter Camera,” presented at the INMM 60th Annual Meeting, Palm Desert, California, USA, Jul. 14, 2019.

= **52 seconds** for ~14 degree resolution, vs our **31 seconds**

10^5 n/s source @ 10 m, with 8 kg of scintillator.

This compares a prototype to our simulation, so premature to claim metamaterial can outperform compact neutron scatter cameras, but final performance should end up close.

But, metamaterial requires 16 sensors for an 8 kg detector. Compact scatter camera requires 512 sensors for 8 kg (and fast timing requirements for sensors). **Potential for significant cost savings**, giving metamaterials a niche even if performance ends up lower.