05-12-2025

Nuclear recoil detection with color centers in bulk lithium fluoride

based on arXiv:2503.20723

3rd workshop on
Mineral Detection of Neutrinos and Dark Matter
JAMSTEC, Yokohama, Japan
May 20-23, 2025

Patrick Huber for the PALEOCCENE collaboration
Center for Neutrino Physics at Virginia Tech

Damage formation

The latent track of crystal damage is amplified by chemical etching (dry or wet)

And then features become large enough to be seen under a conventional microscope.

Standard technique in geo-chronology

These damage features are persistent over geological times!

G. A. Wagner, P. van de Haute, *Fission track dating,* Ferdinand Enke Verlag, 1992

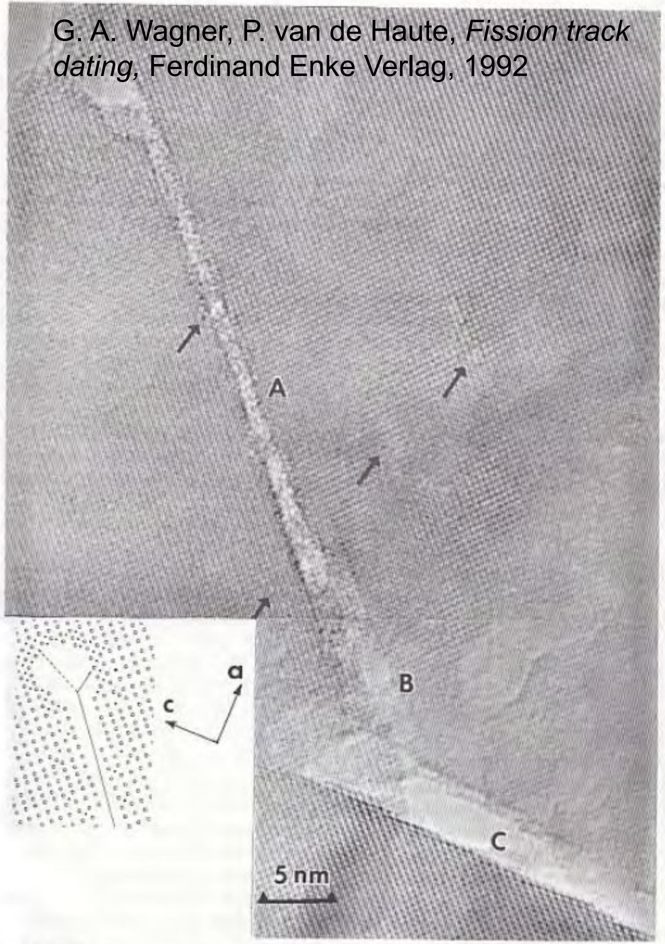
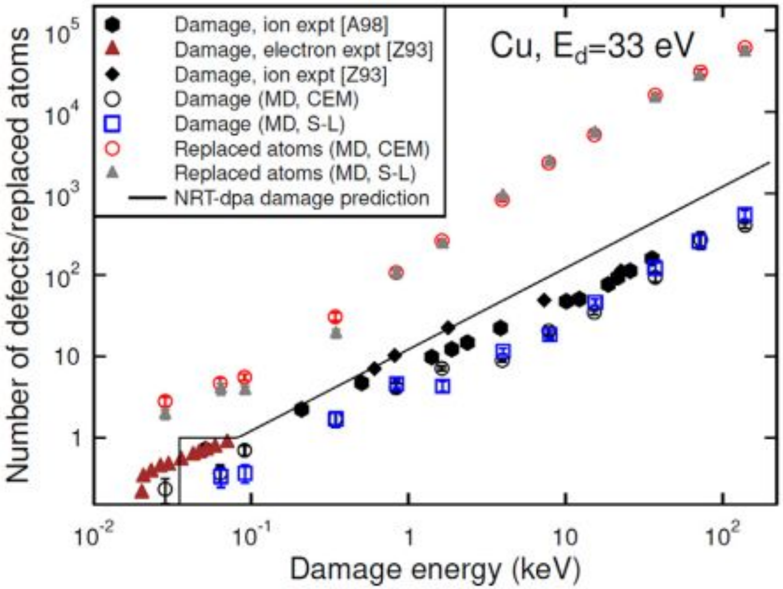


Figure 1.1. Three uranium fission-tracks (A, B and C) making different angles with the (100) plane of zircon (= plane of the picture) observed with high resolution electron microscopy. Arrows indicate sites of point defects. The inset visualizes the formation of the damage track according to Yada *et al.* (1987) together with two major crystallographic directions. (Electron micrograph obtained by courtesy of Dr K. Yada.)

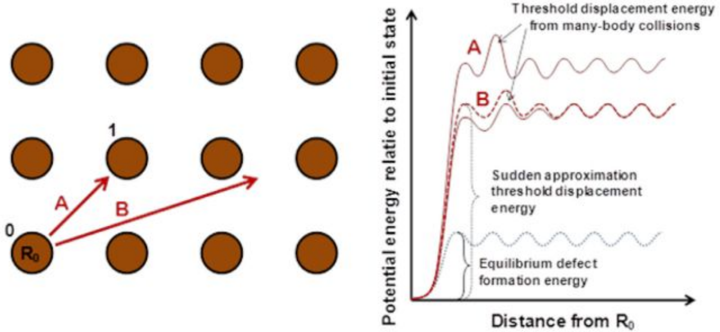
Vacancy formation

- Need big enough separation to prevent recombination
- Main mechanism for radiation damage in e.g. reactor vessels
- We use the TRIM package in full cascade mode

Ziegler, Ziegler, Biersack, NIM B **268** (2010) 1818.

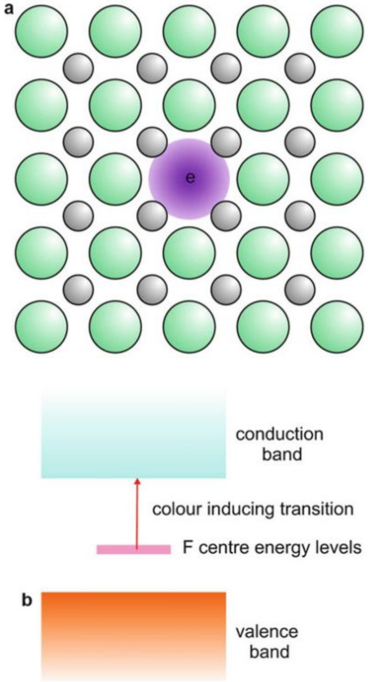


Nordlund, *et al.,* J. Nucl. Mat. **512** (2018) 450-470



Color centers

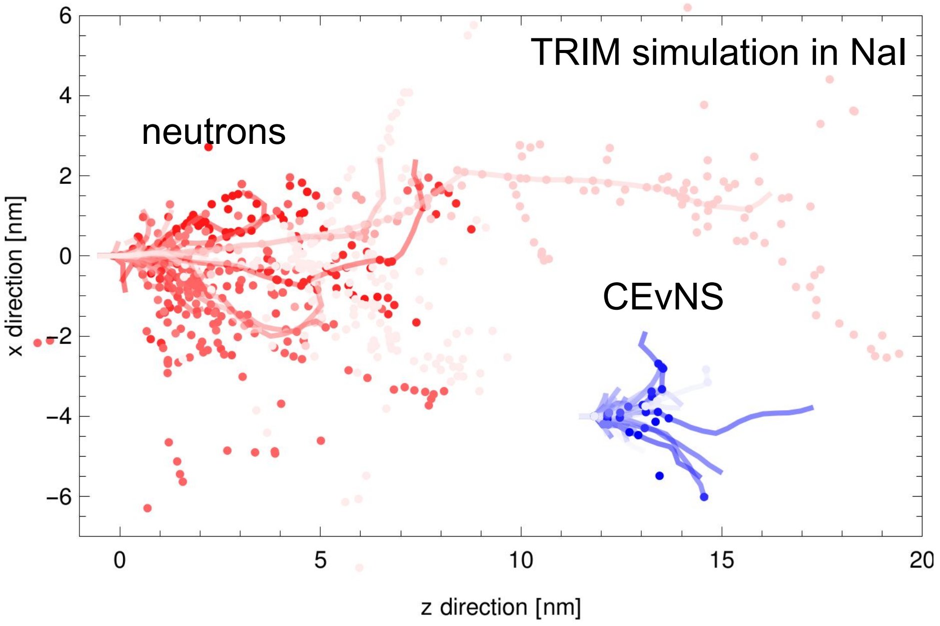
- In ionic crystals (e.g. NaCl), anion vacancies trap electrons
- Quantum mechanics in a square well → distinct energy levels
- Individual color centers can be seen in visible light by fluorescence spectroscopy (NV in diamond, SiC)
- Observed in a wide class of materials



R. Tilley, Encyclopedia of Color Science and Technology, Springer 2013

The concept

- Nuclear recoils damage crystal lattice permanently, either by forming tracks or vacancies
- This allows off-site readout, hence detector is passive
- Intrinsic rejection of ionizing backgrounds, leaves only neutrons
- This has been explored for dark matter and neutrino detection

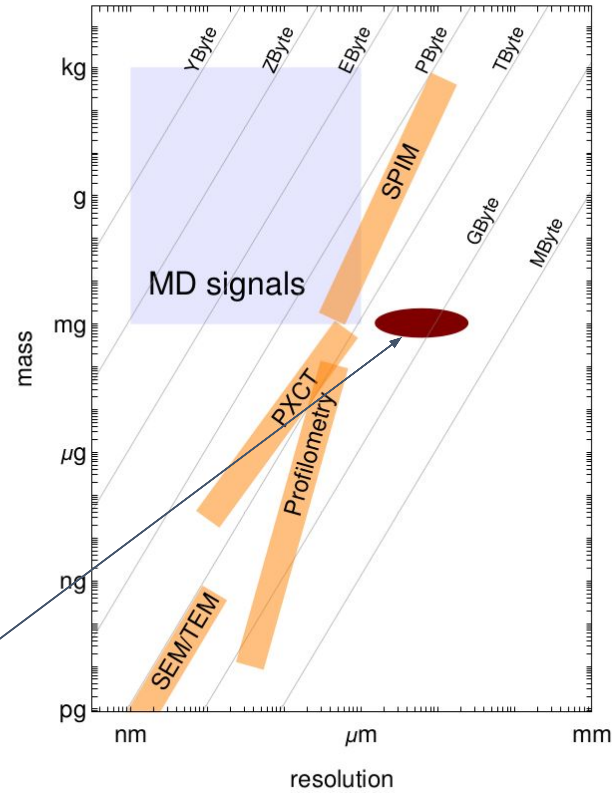


Cogswell, Goel, Huber, Phys. Rev. Applied **16**, 064060

Readout challenges

Radiation induced features are long, narrow and occupy a tiny fraction of the volume:

- SPIM – selective plane illumination microscopy
- PXCT – coherent x-ray imaging
- Profilometry – after etching
- SEM/TEM – scanning/transmission electron microscopy



This corresponds to the data shown in this talk

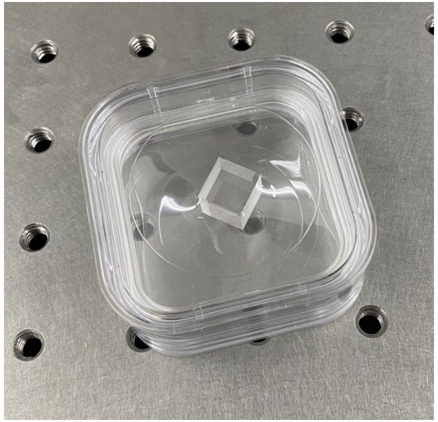
Mass throughput versus resolution, blue box region of interest for mineral detection

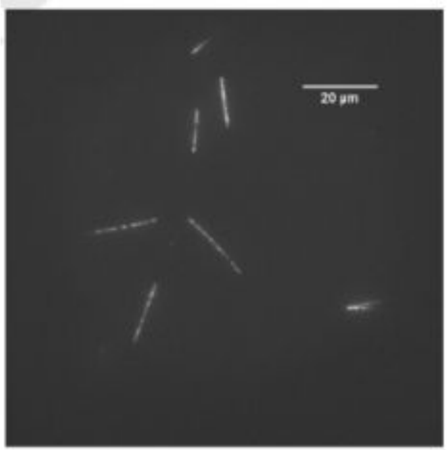
Why lithium fluoride?

- Readily available optical quality crystals
- Previous studies on use as fluorescent nuclear track detector (FNTD): P. Bilski *et al.* have studied many different irradiation modalities
- Visible wavelength for absorption/emission
- ^{6}Li content provides thermal neutron signature

$$^{6}\mathrm{Li} + n \rightarrow {}^{3}\mathrm{H} + \alpha$$

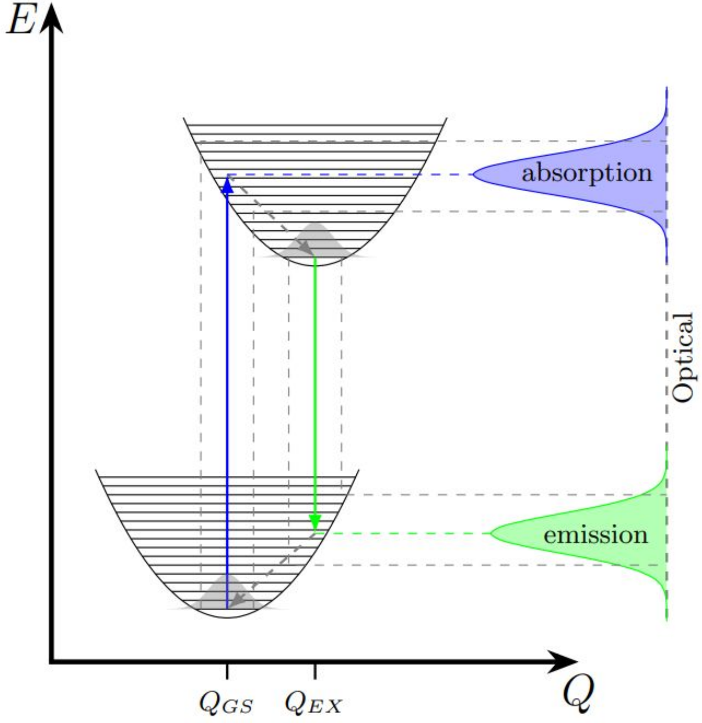
BUT does not occur naturally in useful quantities

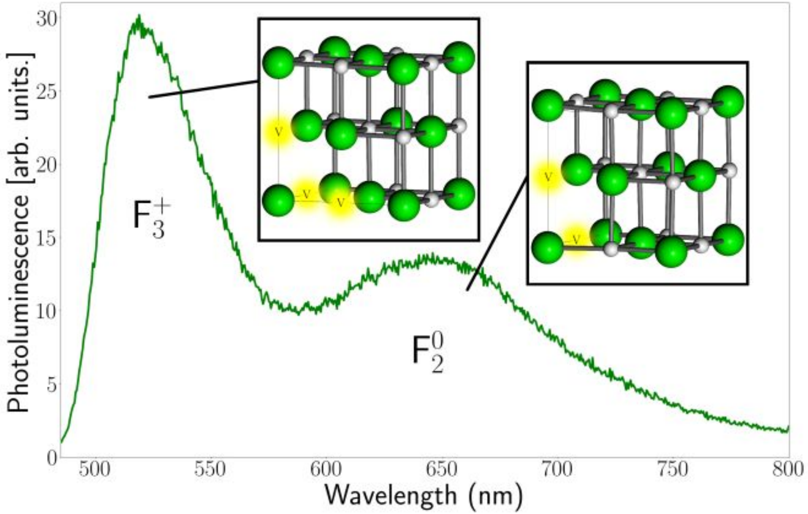




P. Bilski, D. Marczewska, M. Sankowska, W. Gieszczyk, J, Mietelski, Measurement **160** (2020) 107837.

Ab initio theory



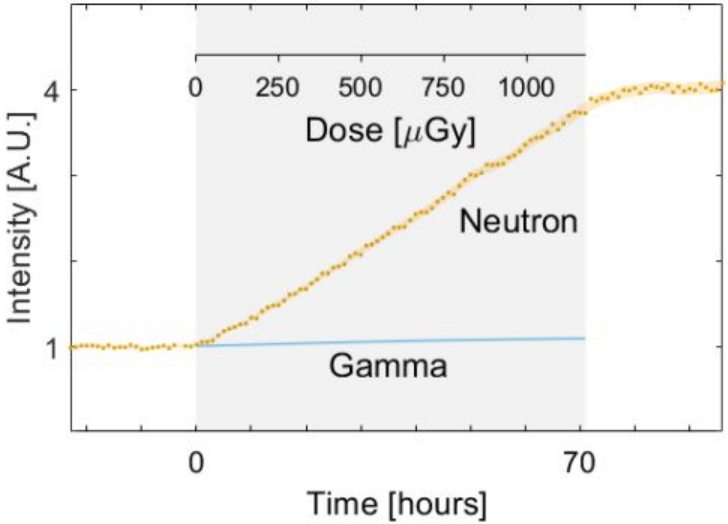


	absorption	emission
F_2^0	500nm (450nm)	650nm (650nm)
F_3^+	440nm (450nm)	470nm (530nm)

M. Guerrero Perez, K. Walkup, J. Chapman, P. Bhaumik, G.A. Khodaparast, B.A. Magill, P. Huber, V. Ivanov, arXiv:2412.21060

Bulk fluorescence

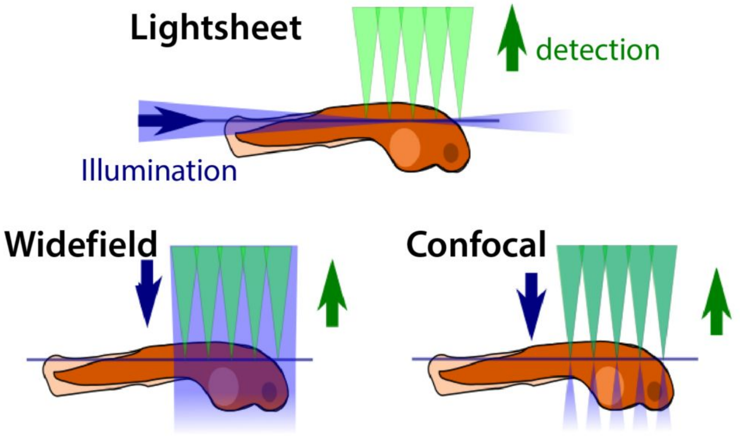
- AmBe neutron and ^{60}Co gamma source, dose rate determined from GEANT4 simulation
- *In situ* measurement of photoluminescence using cold SiPMs (single photon counting)
- Dose-for-dose 50 times less sensitivity to gammas than to neutrons



3D microscopy

SPIM – Selective Plane Illumination Microscopy

- SPIM aka lightsheet microscopy is well known technology in biology
- Confocal microscope-like resolution and sensitivity at 10 million times higher throughput
- Compact systems



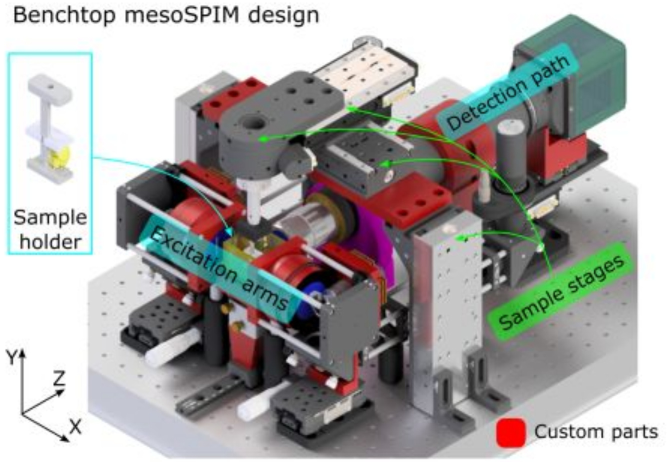
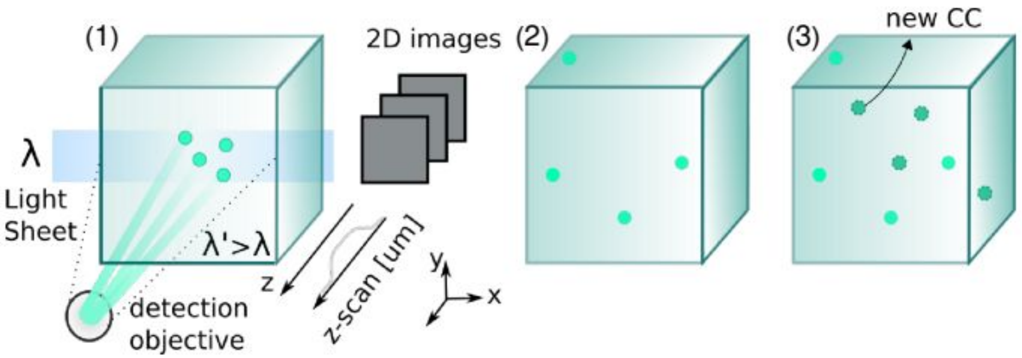


Figure courtesy N. Vladimirov, U. Zurich

SPIM



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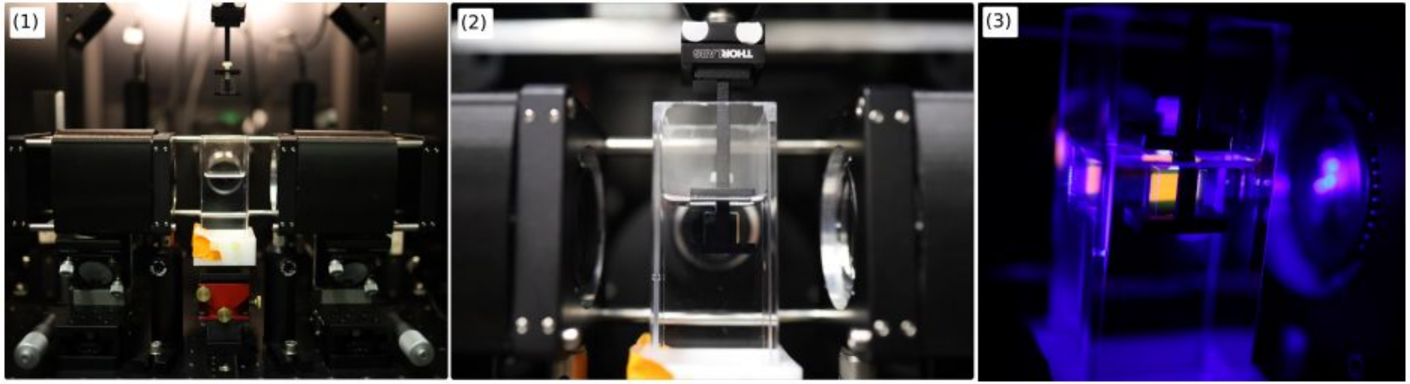

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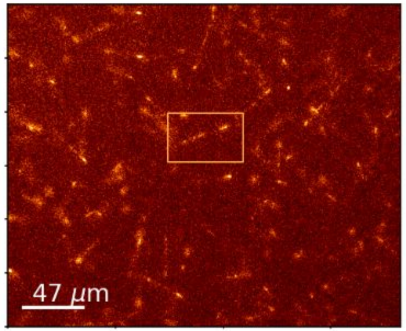
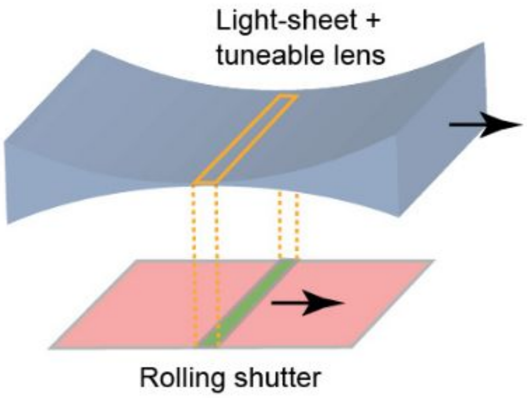
THORLABS
(3)

Seeing tracks for real



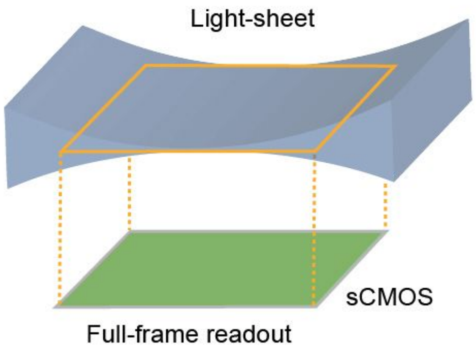
Need lots of light to get above noise.

To achieve uniform z-resolution use axially swept light sheet (ASLM).



Bleaching (photo ionization) limits how much laser light we can use

- → need to turn of ASLM
- → long exposures
- → **slow scan speed**



non-uniform z-resolution!

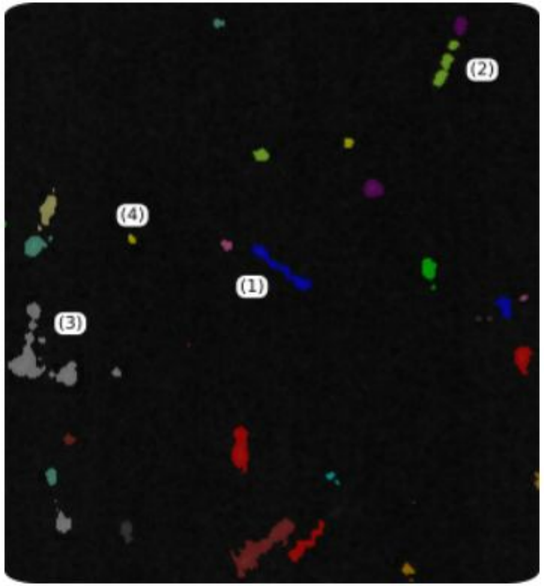
schematics from mesospim.org

Data analysis

0504	0505	0506	0506
Thermal neutrons	Fast neutrons	Cosmics only	Comics only
Room temp	Room temp	Room temp	6 hours at 350C

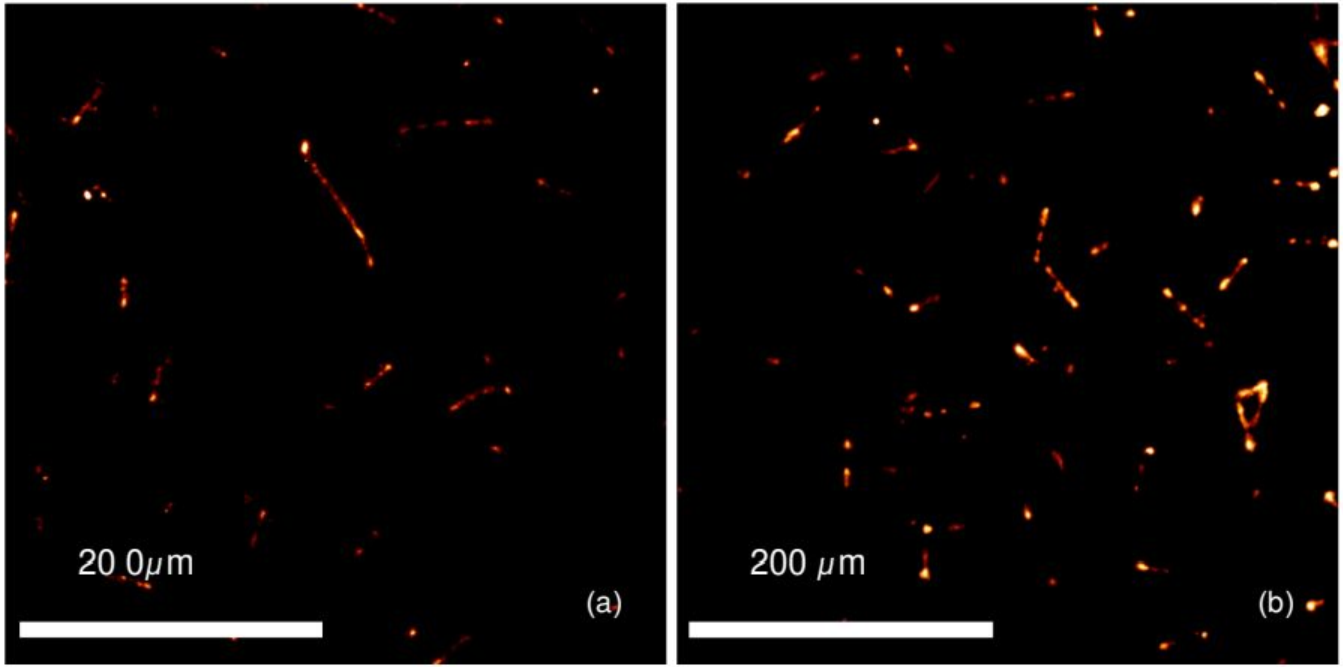
1) Pre-processing and de-noising
2) Segmentation and feature identification (ML-based)
3) Feature size-based selection
4) Rejection of mis-formed events (human)

Imaged regions ~0.2x0.2x0.1mm³, 1-2mm inside the crystal. Unless otherwise noted, 2D projections shown.



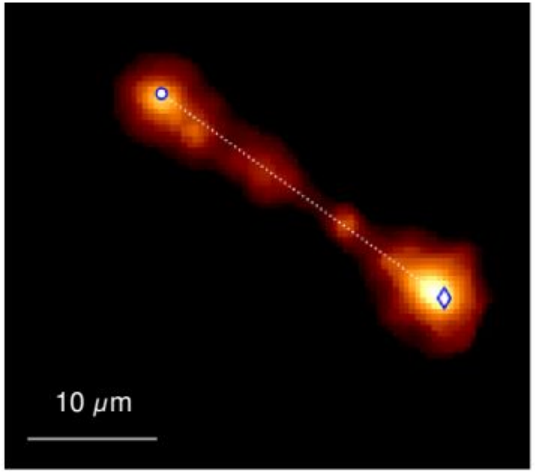
2D projection of segmented data and labeling in *ilastik*

Cosmic rays vs thermal neutrons

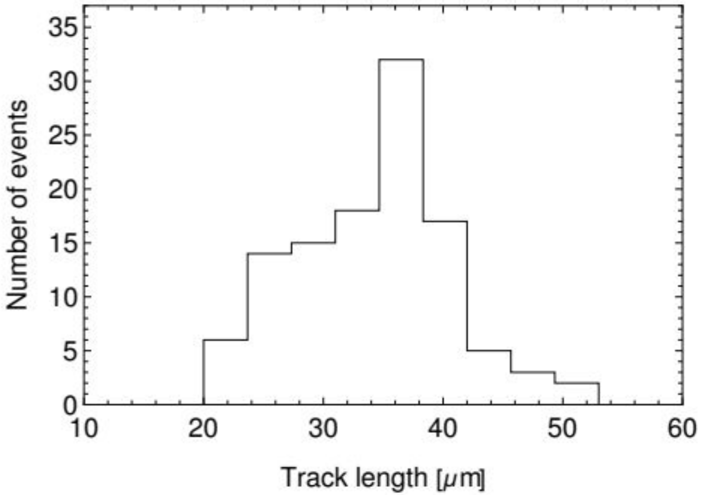


a) 0506 b) 0505

Thermal neutrons track length

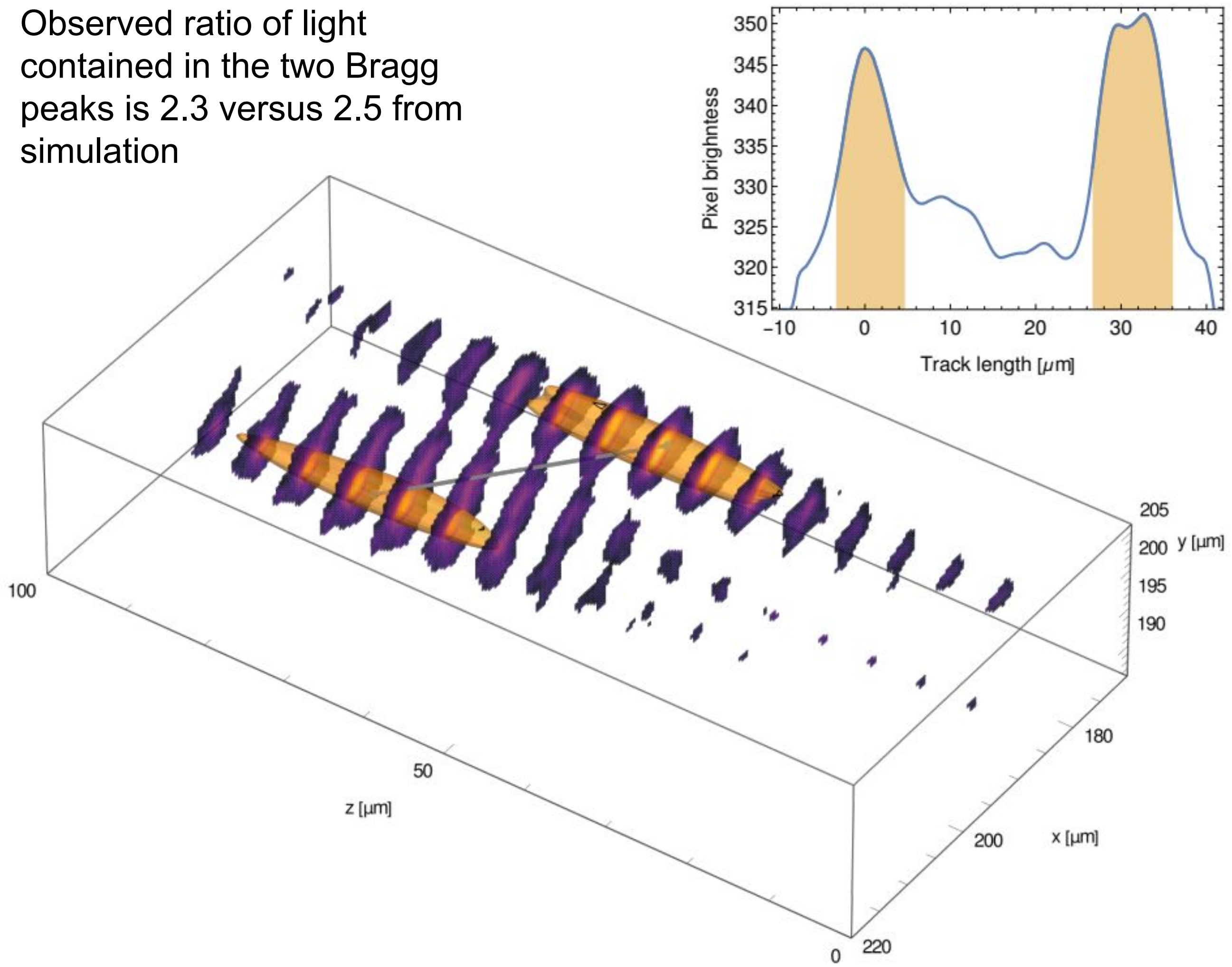


$$^{6}\text{Li} + n \rightarrow {}^{3}\text{H} + \alpha$$



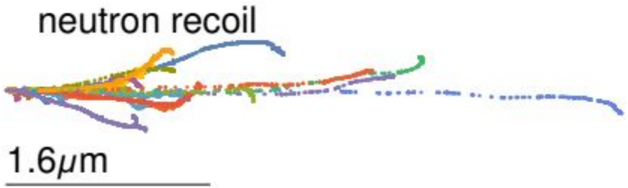
Predicted track length ~35μm

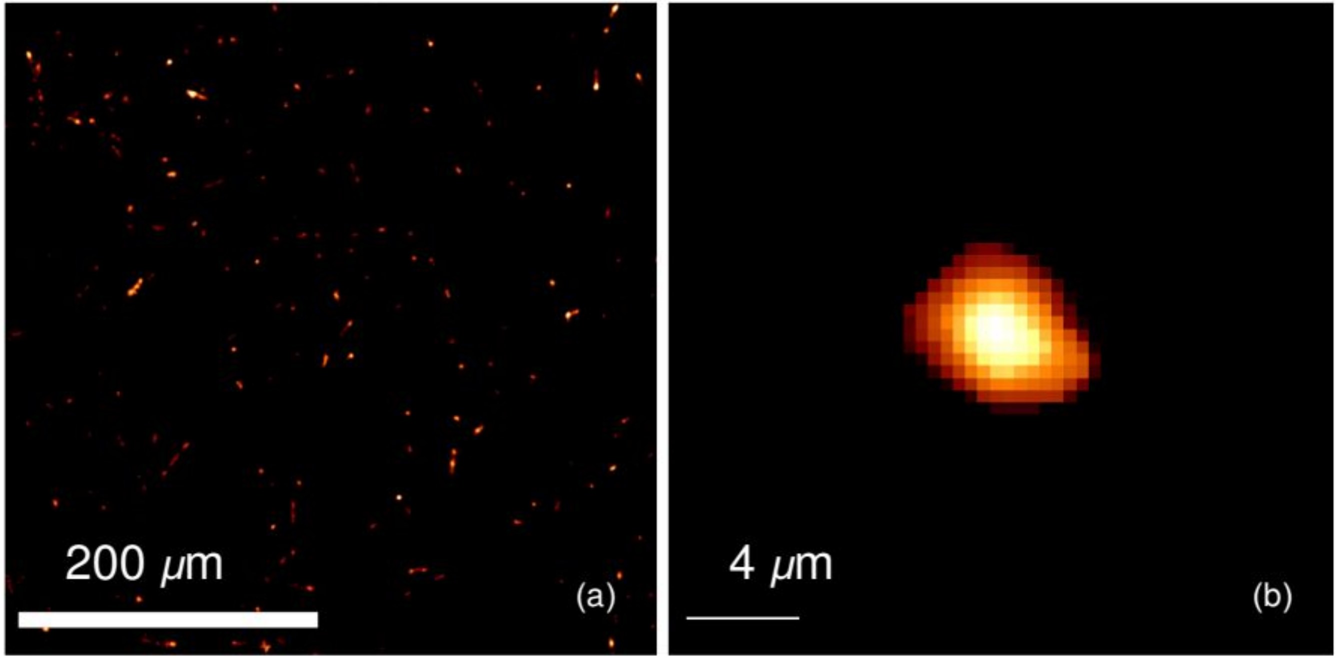
Observed ratio of light contained in the two Bragg peaks is 2.3 versus 2.5 from simulation



relatively low spatial resolution in z-direction since we have to turn off ASLM

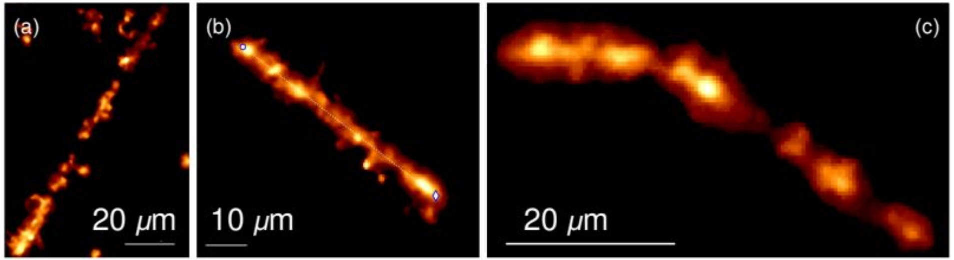
Fast neutrons

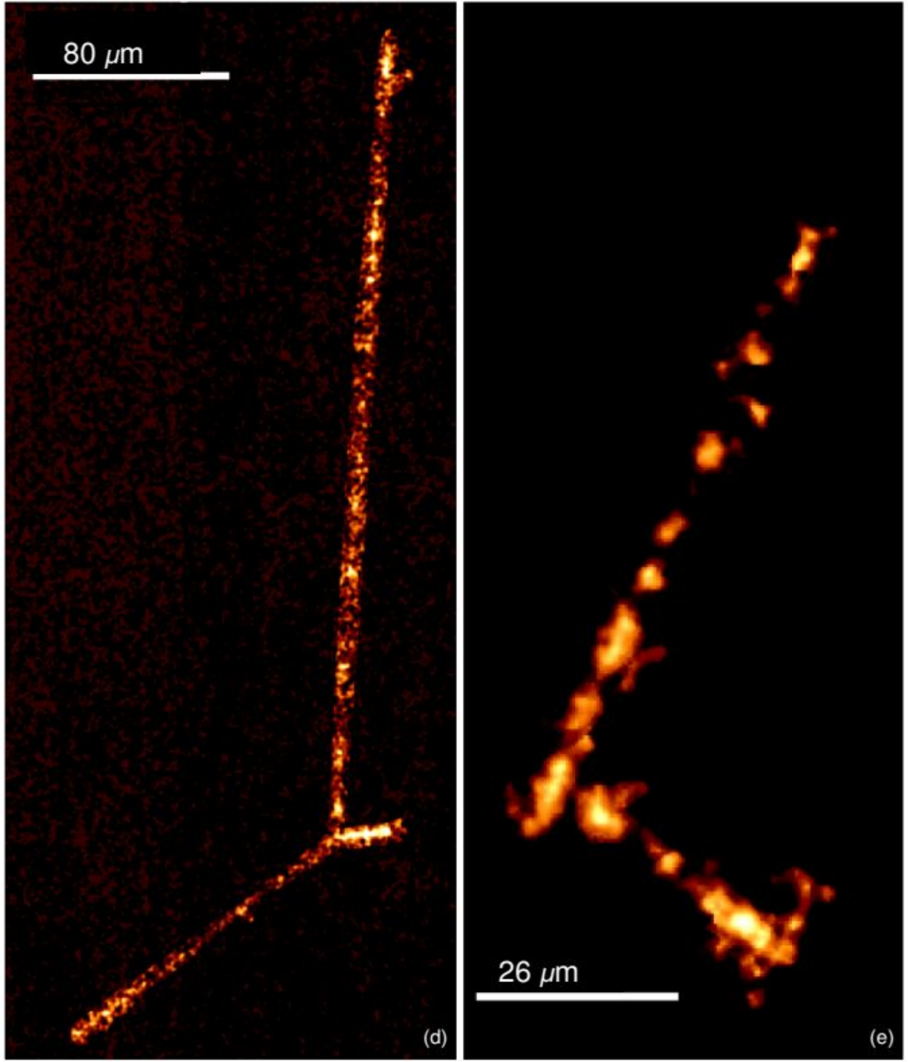




Cosmic rays

We observe a number of large multi-track events and assume they are from energetic cosmic ray interactions, e.g. muon spallation.

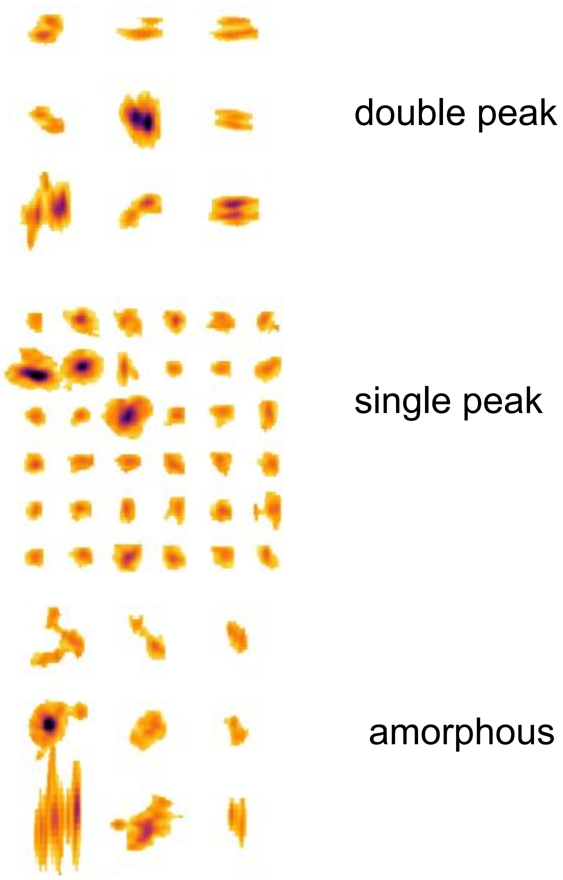




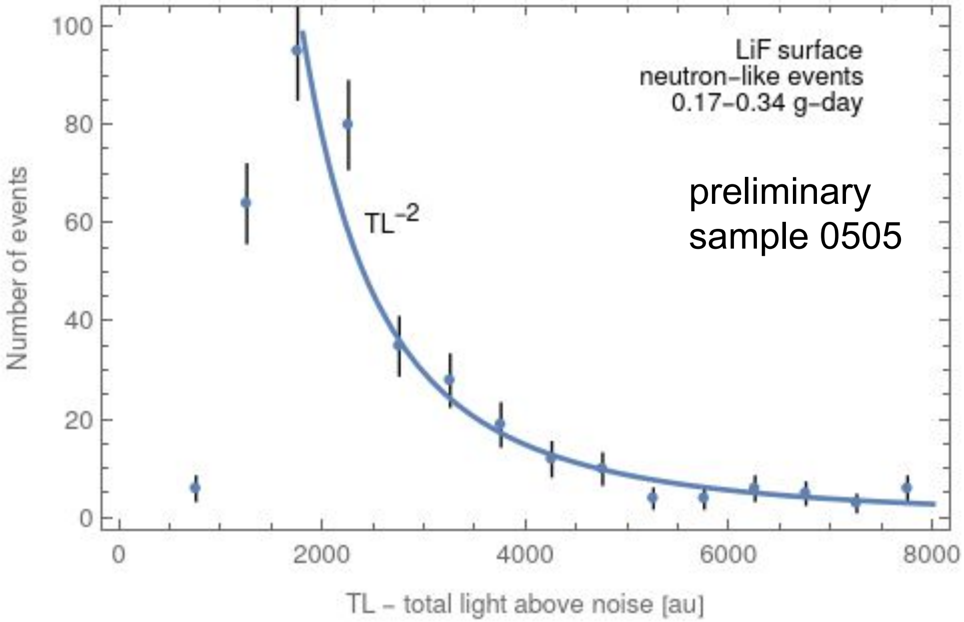
Event identification

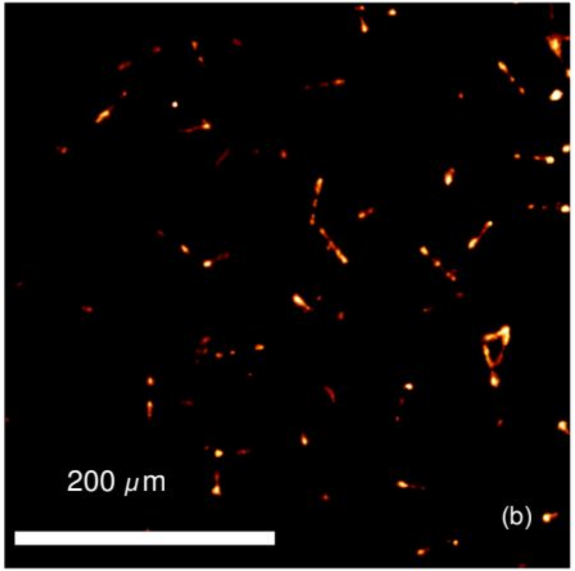
Hand-scan of 0.18mm³ in sample 0505

	predicted	observed
Thermal neutron (double peak)	100-160	136
Fast neutron-like (single peak)	100-600 (mostly from cosmic rays)	422
Amorphous	n/a	385



Neutron background data

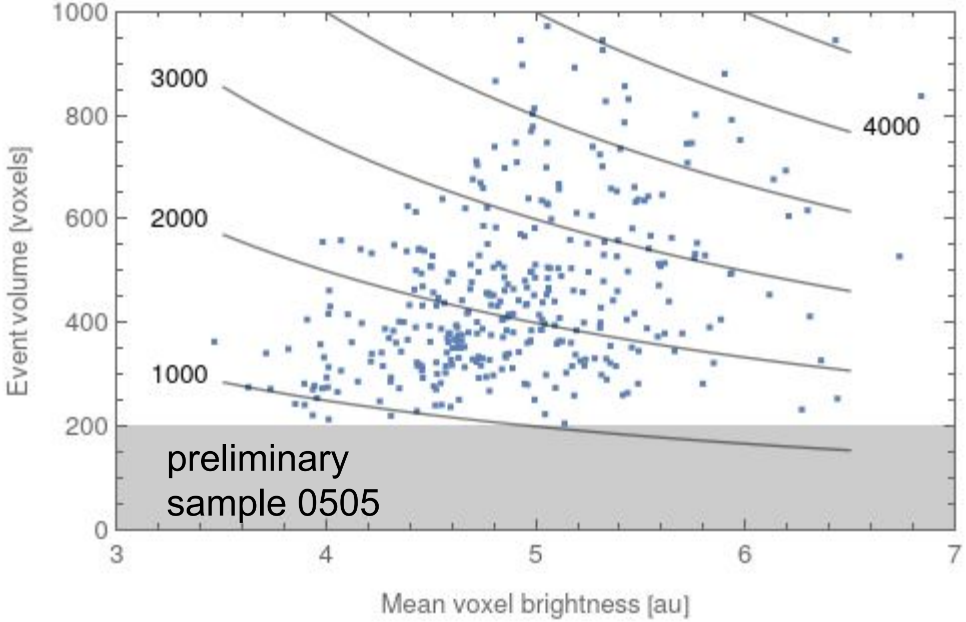




This corresponds to about 10^6 events per kg day

Total light (TL) is a measure of the number of color centers

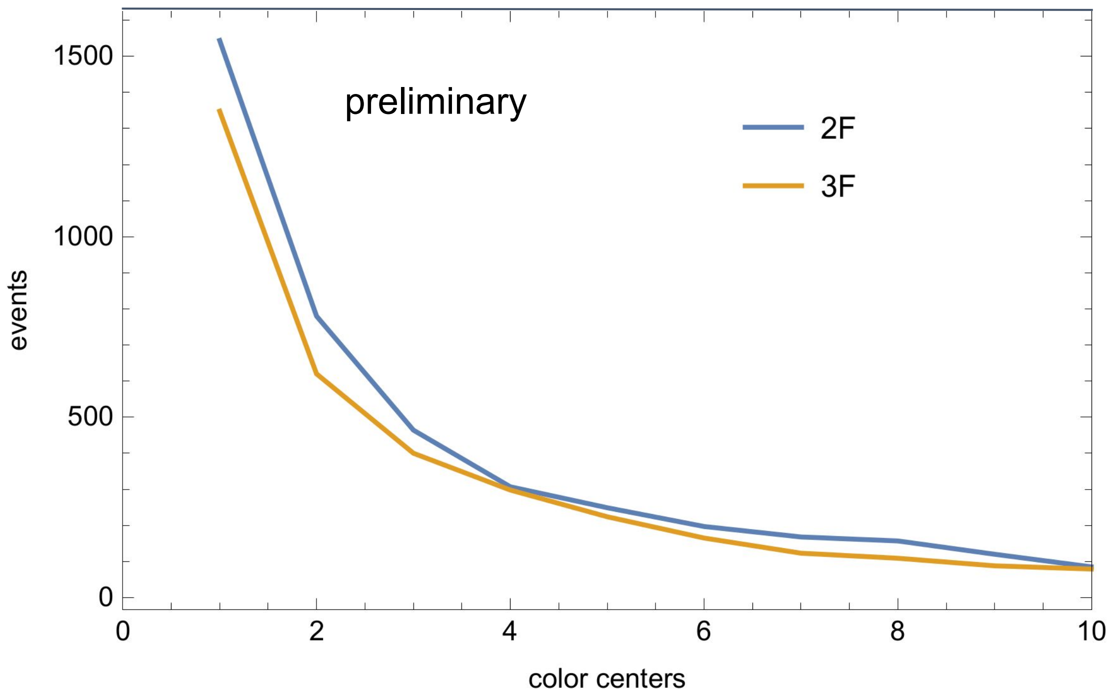
Neutron background data



Resolution limits the smallest object we can image to about 200 voxels.

Low energy drop in TL distribution probably due to minimum resolvable volume and not brightness.

Neutron background simulation



Neutron flux $E^{-1.5}$
elastic neutron scattering only.

Neutron recoils simulated with TRIM with full tracking of secondary cascades.

Color center spectrum scales as power law with index -1.5

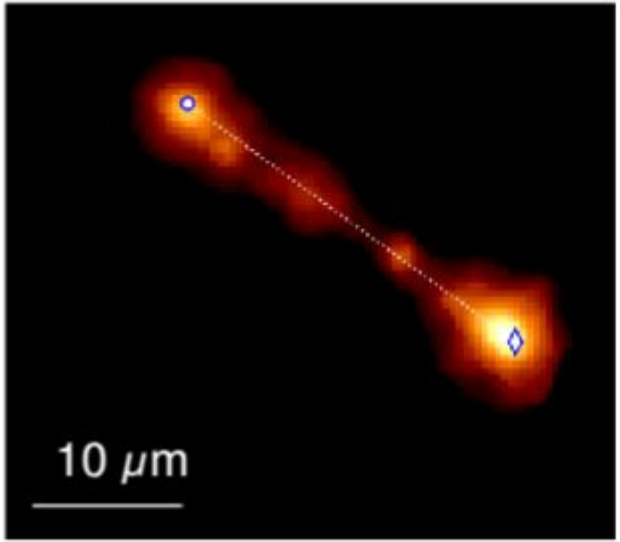
Triton simulation

According to TRIM

- In the Bragg peak we have about 112 F centers/micrometer (FCMM)
- The next brightest pixel has 30 FCMM
- The mean number of FCMM outside of the Bragg peak is less than 10
- The mean of the minimum brightness along the track is 3 FCCM

We have to divide these number by 2 or 3 to obtain the number of optically active F2/F3 centers.

Triton data – low recoil energy threshold



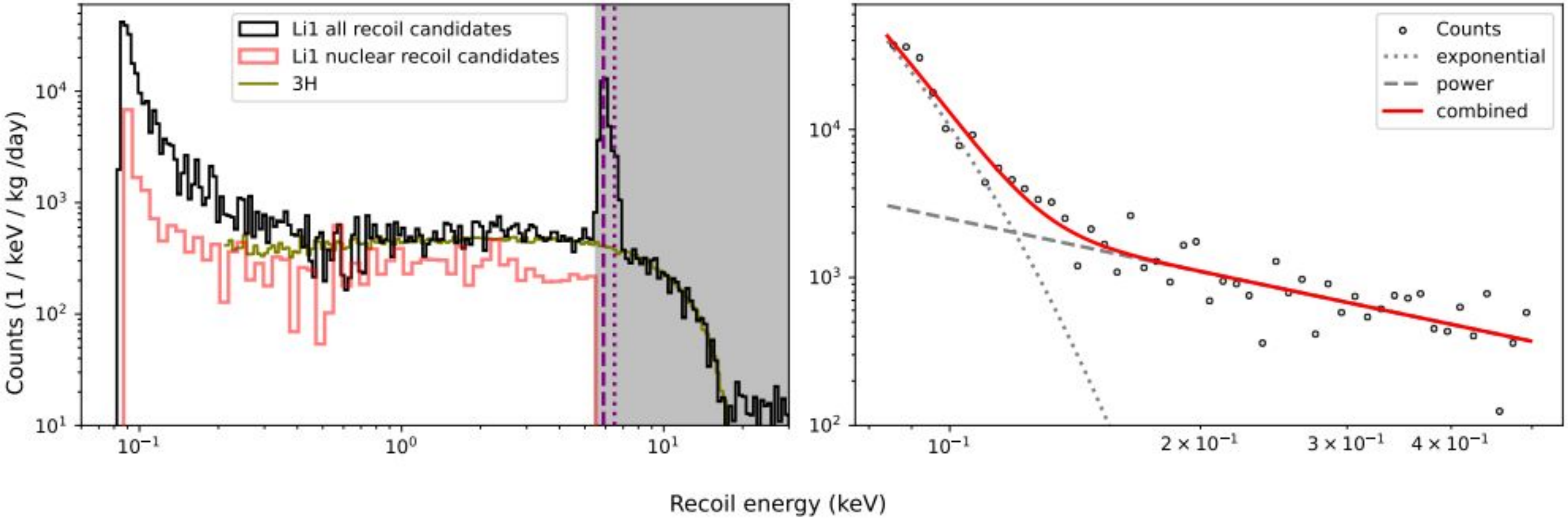
We see the track of the triton in most cases!

- With high confidence we see pixel with 3-5 color centers
- With medium confidence we see pixel with 1-2 color centers

And this is with the current camera noise level.

CRESST LiAlO2 data

arXiv:2207.07640



Power law index 1.2, total rate about 1300 events/kg/day/keV.

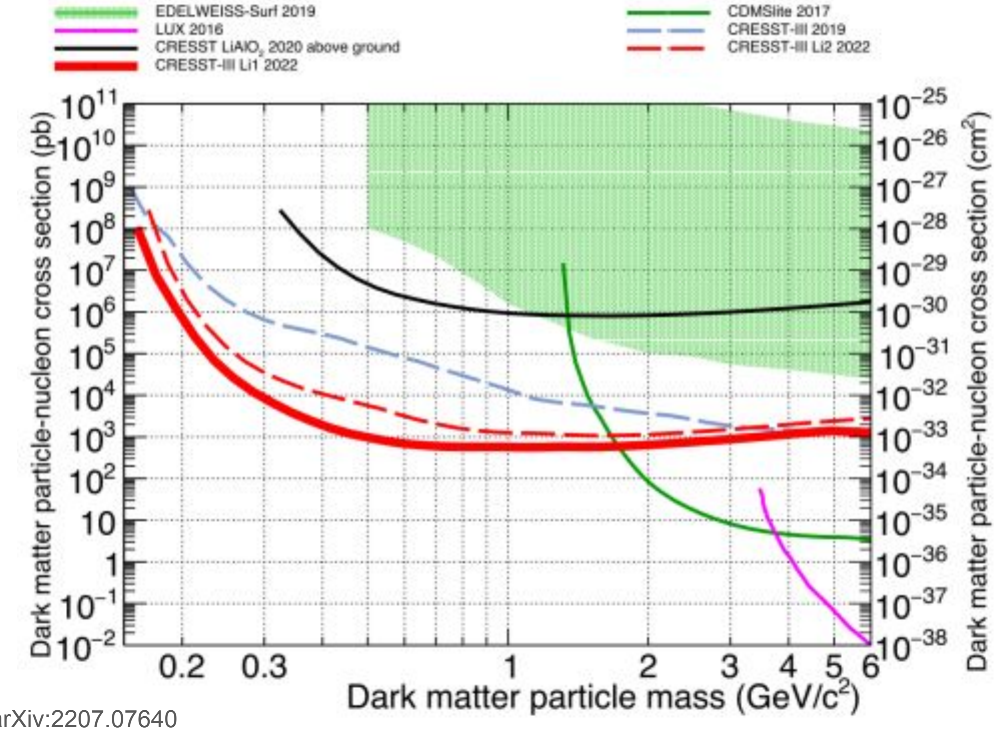
arXiv:2005.02692 reports a surface rate of $2x10^5$ events/kg/day/keV

CRESST LiAlO2 data

- 1.2 kg-days exposure
- Recoil threshold 83.6eV
- Background limited
- Going from surface to underground reduced background by 150x
- Fast neutron flux in Gran Sasso is about 10^6 times lower than at the surface

EPJC **79** 747 (2019)

Neutron

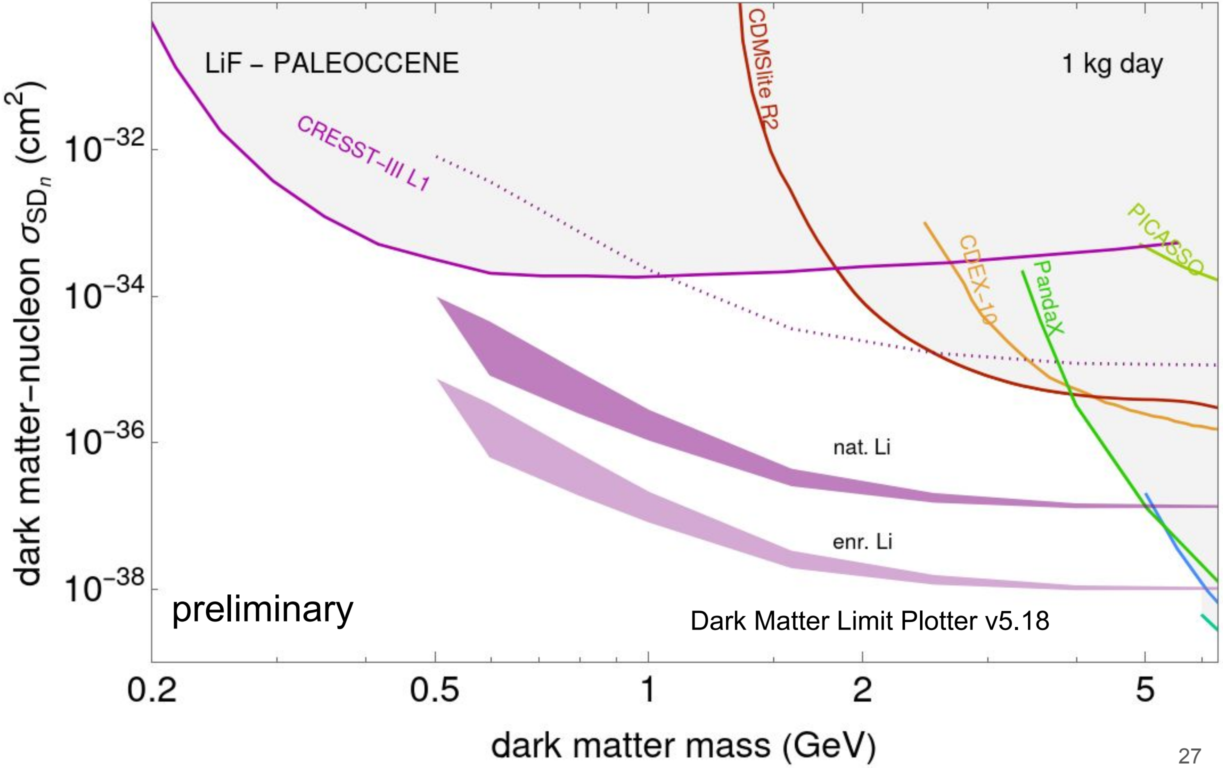


arXiv:2207.07640

Direct DM search with lithium fluoride

- TRIM simulation
- Effective 200-300eV recoil threshold
- Our observed background scaled by 10^6
- With a $1cm^3$ target we need 380 days of exposure and 9 days of scanning (at current speed)

dotted line - our observed background scaled by 150



Reactor CEvNS

LiF not ideal because of low A, but low mass implies higher recoil energies

CEνNS event numbers are computed for 100 g of target and 1-year exposure at 10 m from a 3-GW_{th} reactor. The same number for inverse beta decay on CH_2 is 1250. The selection efficiency is the fraction of reactor CEνNS events that have one or more vacancies created. The usable events are the number of events where one or more vacancies are created.

Material	A (u)	m_A (%)	Density (g cm^{-3})	Melting point (K)	TDE (eV)	$E_{1/2}$ (eV)	CEνNS events	Selection efficiency (%)	Usable events
LiF	19.0	73.2	2.64	1 120	27	80	5 600	75	4 200
BaF_2	137.3	78.3	4.88	1 625	35	105	48 800	20	9 600
NaI	126.9	85.0	3.67	935	24	65	46 900	32	15 100
CsI	132.9	~ 100	4.51	900	23	55	55 700	37	20 600
$CaWO_4$	183.8	63.9	6.06	1 895	41	110	55 400	8	4 600
$Bi_{12}GeO_{20}$	209.0	86.4	9.22	1 175	28	85	83 500	17	14 000

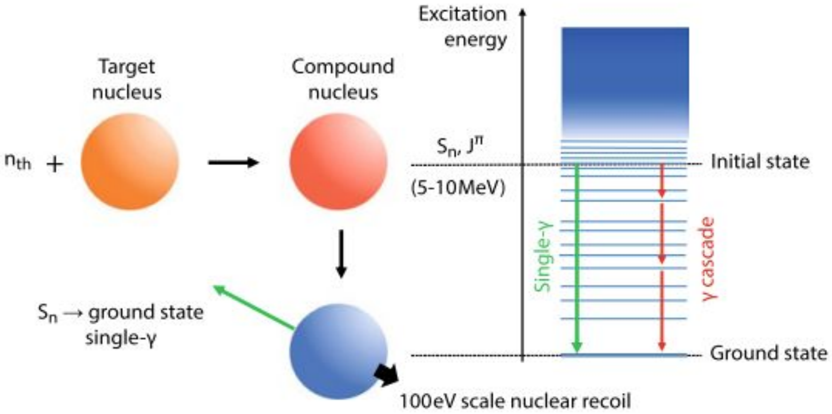
Cogswell, Goel, Huber, Phys. Rev. Applied **16**, 064060

2F-3F	6F-9F	20F-30F
3389-2834	1771-1165	294-89

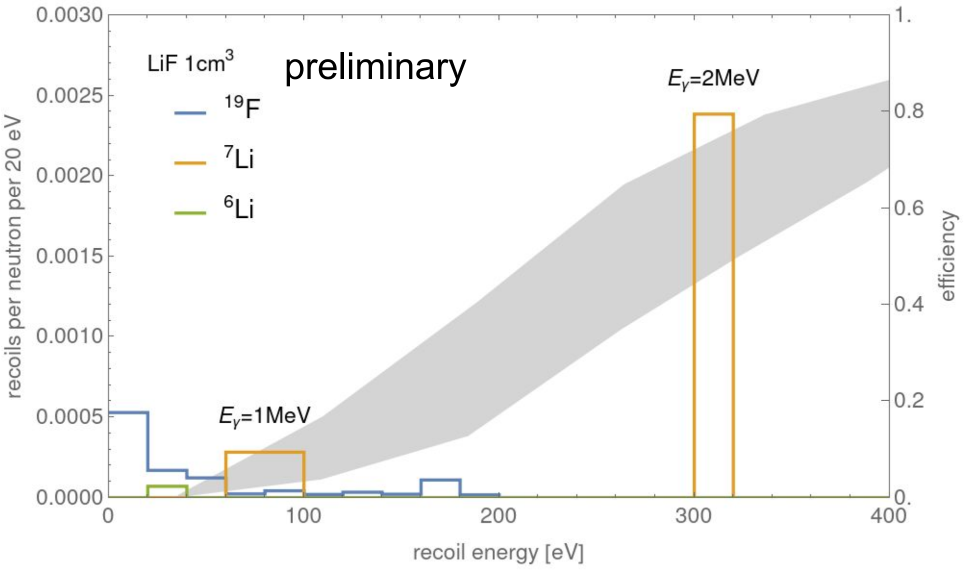
About 3000 events per 100g yr but surface background is $5x10^5$… With a factor 100 from passive shielding and a factor 3 from the spectrum still get a 8-10σ significance (no systematics)

- **Need to understand low-energy response**
- **Need to understand background rate and spectrum**

CRAB-ing



H. Kluck, *et al.* J Low Temp Phys **218**, 101–109 (2025).



Rate depends on capture cross section, isotope abundance and gamma emission probability, energy is driven by gamma energy.

Measuring the gamma energy with a backing detector may allow for detailed response studies.
Figure based on data from https://www.nndc.bnl.gov/capgam/

CRAB - Calibration by Recoils for Accurate Bolometry Phys. Rev. Lett. **130**, 211802 (2023)

Summary

- Gamma/neutron luminescence/dose-rate relationship measured in bulk
- Lithium fluoride shown to be robust against gamma radiation
- Shown persistence of luminescence at room temperature over many months
- 3D imaging of tracks using the mesoSPIM
- ML-based feature identification
- Clear event identification via size and topology
- dE/dx-signatures observed
- Low-LET triton tracks clearly visible
- Thermal and fast neutron rates match expectation

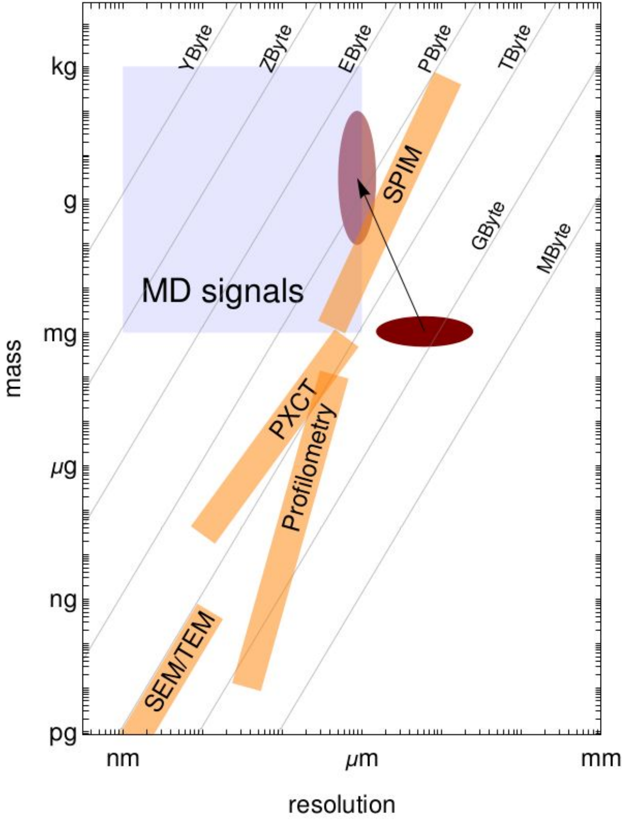
Together with the GCR MDDM significant effort at simulation/reconstruction.

Next steps

- Building mesoSPIM at VT (long lead-time parts ordered)
- Will use a qCMOS camera with 10x lower noise
  - Faster imaging
  - Lower laser intensity, less bleaching
  - Enables ASLM – improved z-resolution
  - Single color center sensitivity
- Dedicated to nuclear recoil imaging - can tinker at will
- Planning CRAB-style measurement to characterize low-energy response
- Looking into options for underground & reactor deployments to characterize backgrounds

We expect to reach scan speeds of order cm³/hour.

Together with the GCR MDDM looking also into other materials that could serves as targets for paleo-detection.



The PALEOCCENE collaboration

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NSF GCR Mineral Detection of Dark Matter

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