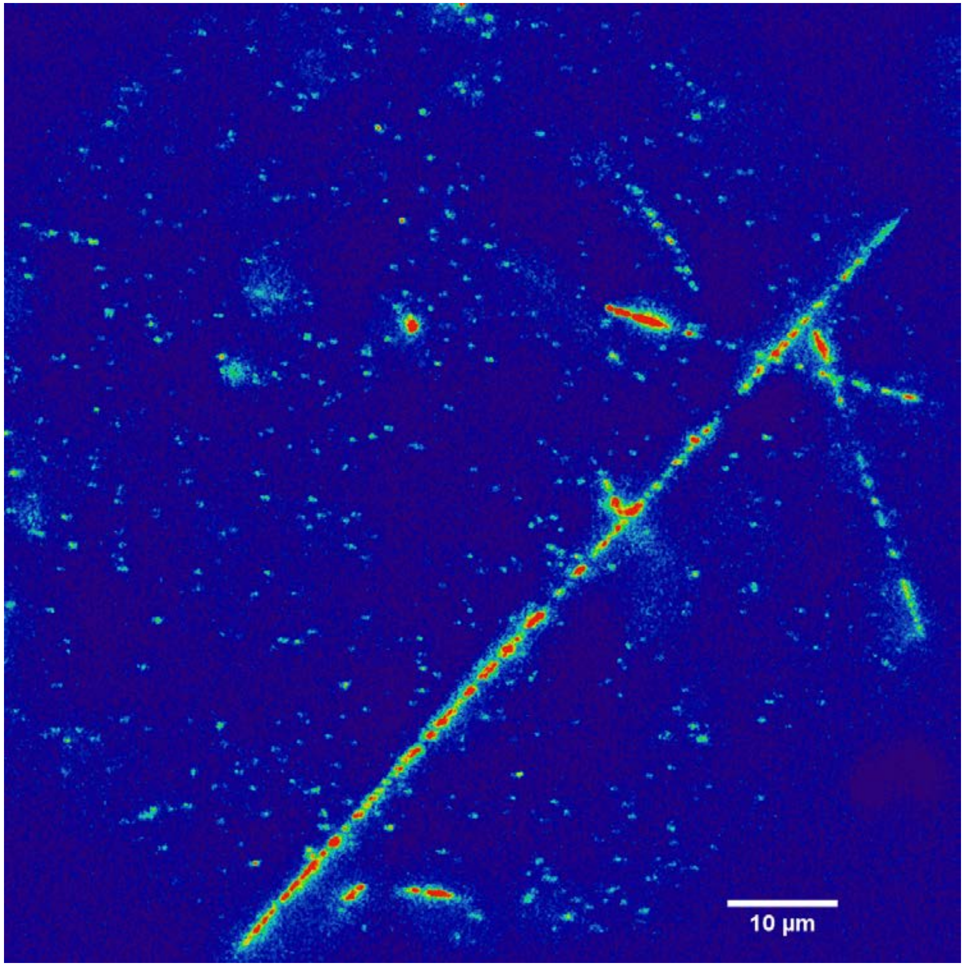
Fluorescent Nuclear Track Detectors (FNTD) based on LiF single crystals

P.Bilski[1], B.Marczewska[1], W.Gieszczyk[1], M.Kłosowski[1], M. Sankowska[1], S.Kodaira[2], B. Przybyla[3], T. Berger[3]

1- Institute of Nuclear Physics Polish Academy of Sciences, Kraków, Poland
2 - National Institute of Radiological Sciences (NIRS) / QST, Chiba, Japan
3 - German Aerospace Center (DLR), Köln, Germany

Fluorescent Nuclear Track Detectors (FNTD)

Examples of **fluorescent tracks** registered in a LiF crystal after 6 months on ISS in frame of DOSIS 3D project

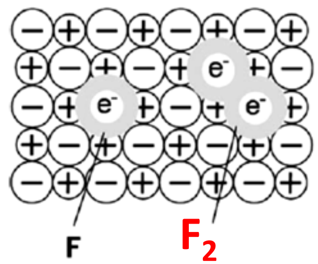


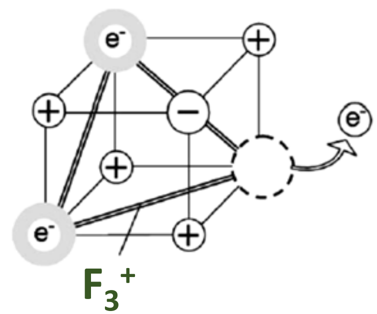
How does it work?:

Physical mechanism: radiophotoluminescence (RPL)

- charged particles create on their path defects (vacancies) in crystal lattice
- defects tend to cluster and trap electrons

in LiF the most interesting are F2 and F3+ defects:





F_2 : 2 anion vacancies + 2 electrons

F_3^+ : 3 anion vacancies + 2 electrons

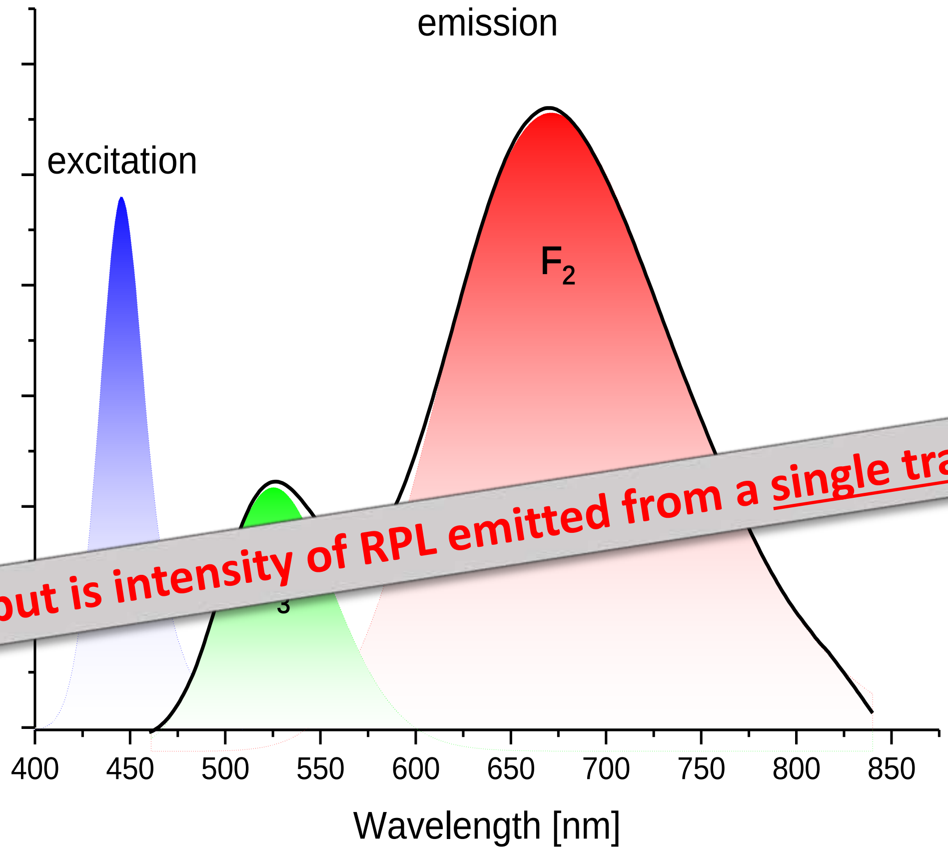
F_2 /F_3^+ are intrinsic defects: pure LiF is used, no need for any activators

- Photoluminescence of F_2 /F_3^+ centers:

<u>upon excitation with **blue** light, **green** and **red** luminescence is emitted</u>

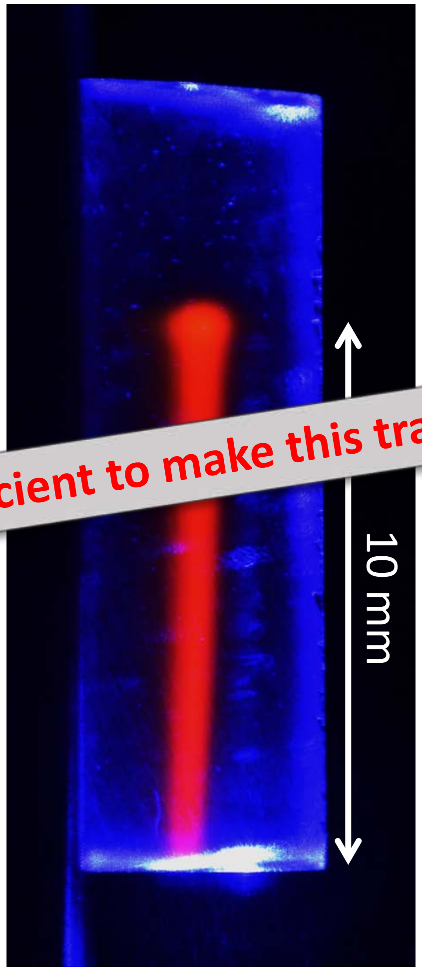
How does it work?:

Upon excitation with **blue** light, **green** and **red** luminescence is emitted



Lithium fluoride photoluminescence spectra

Image of a 50 MeV proton beam stopping in LiF crystal

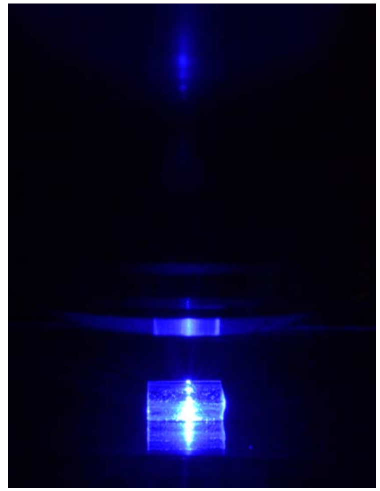


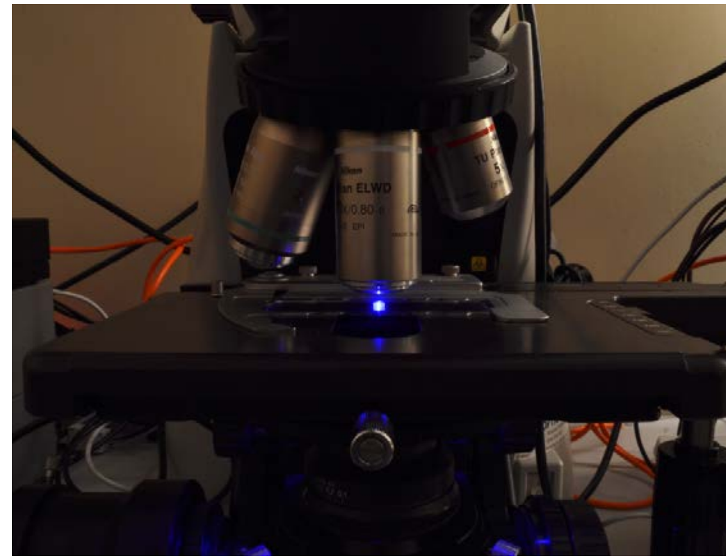
An image of a crystal illuminated with a blue light, taken with a simple photo camera (no filters)

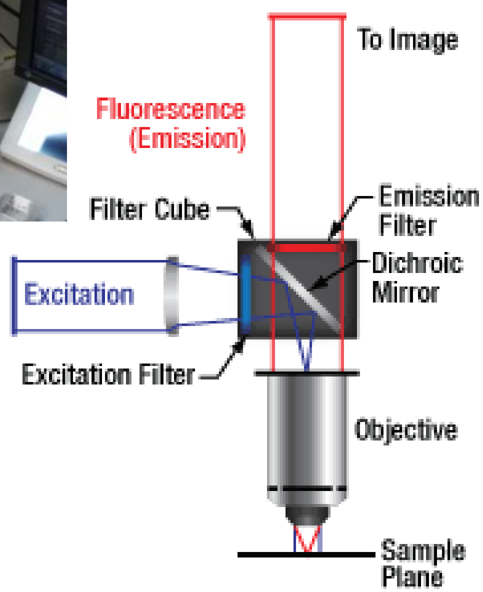
… but is intensity of RPL emitted from a single track sufficient to make this track visible?

Experimental technique: wide-field fluorescent microscope with high-sensitive CCD camera

Nikon Ni-U







Objective: 100×, NA=0.8

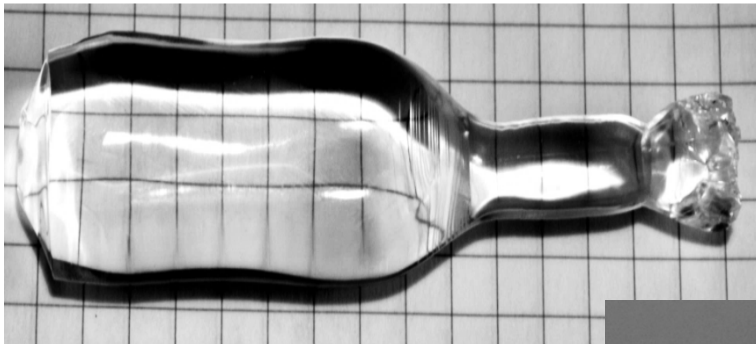
Pixel size: 0.07μm or 0.03μm

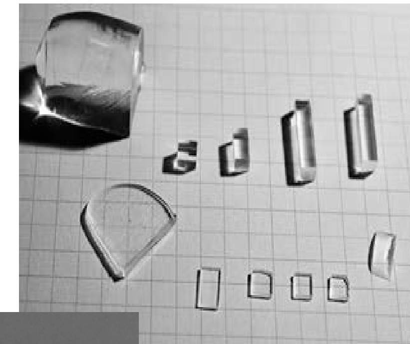
Resolution limit: $\lambda/NA \approx 400$nm

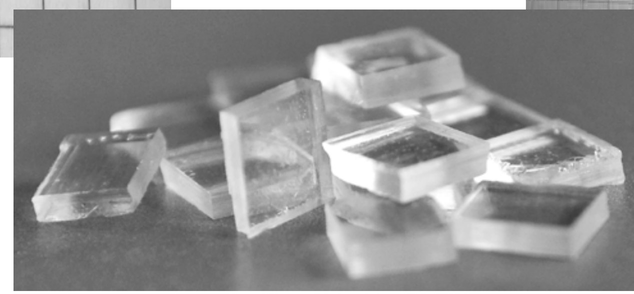
Aquisition time (typical): 1- 60 s

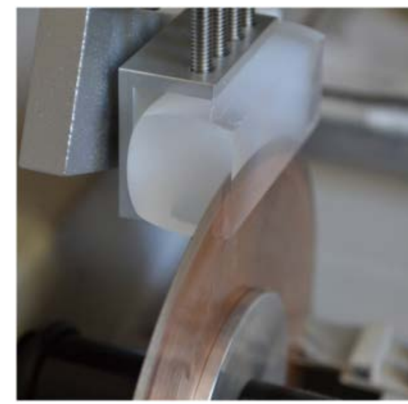
LiF crystals

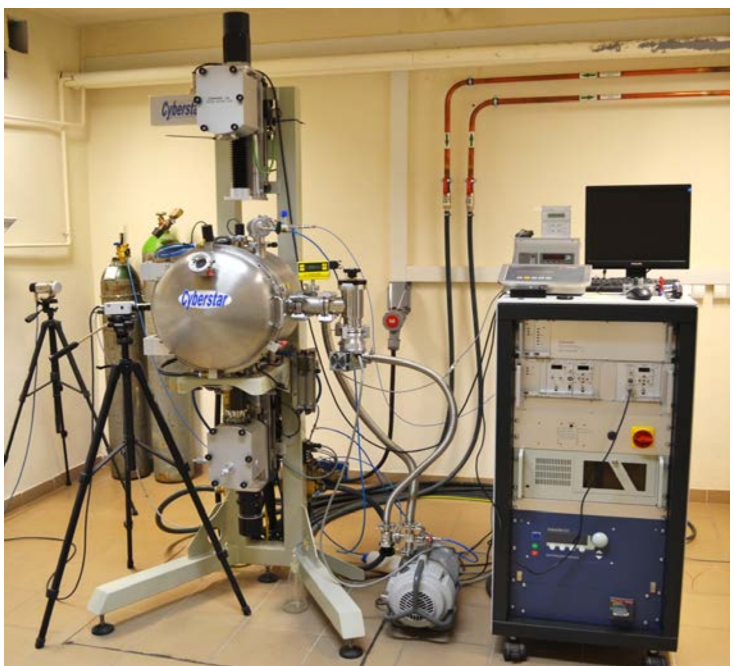
Grown at the IFJ Kraków with the Czochralski and micro-pulling-down methods

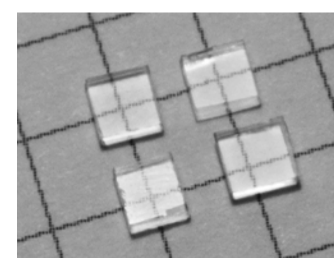




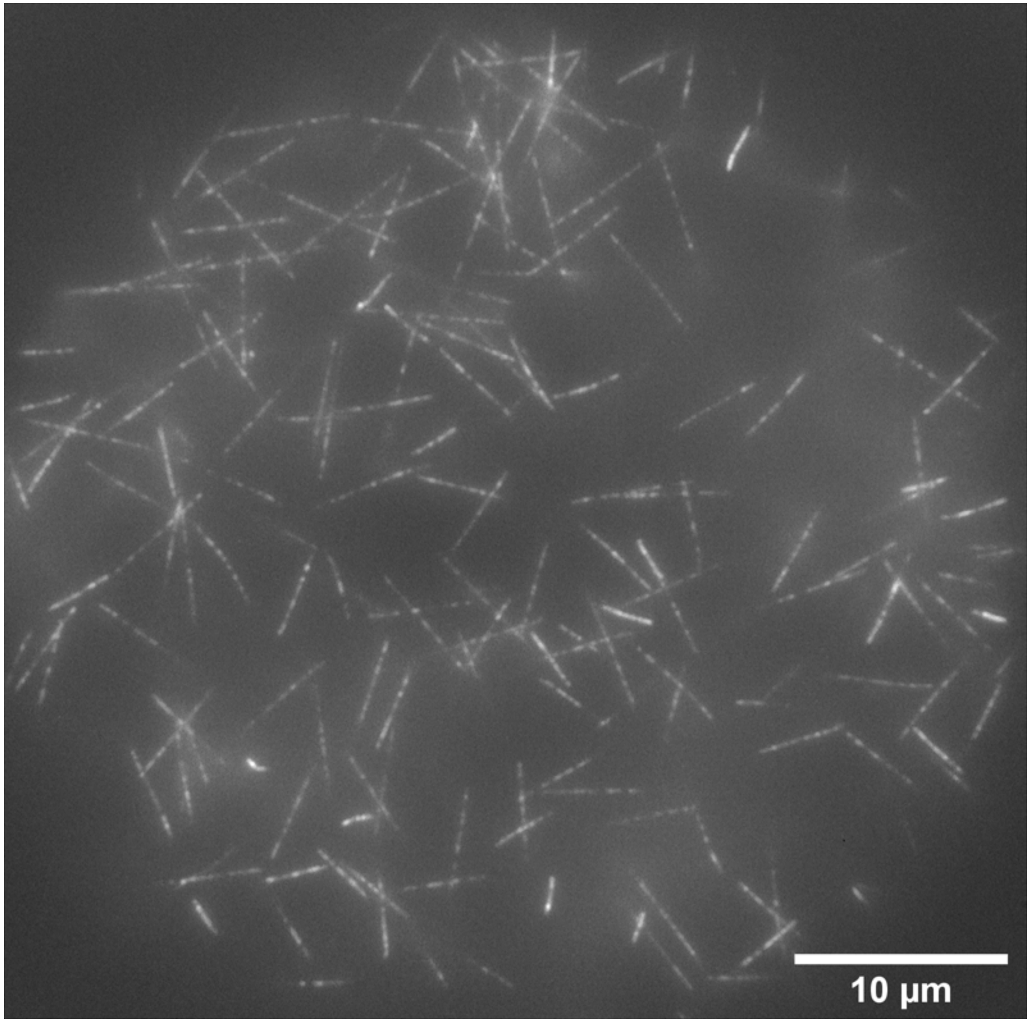






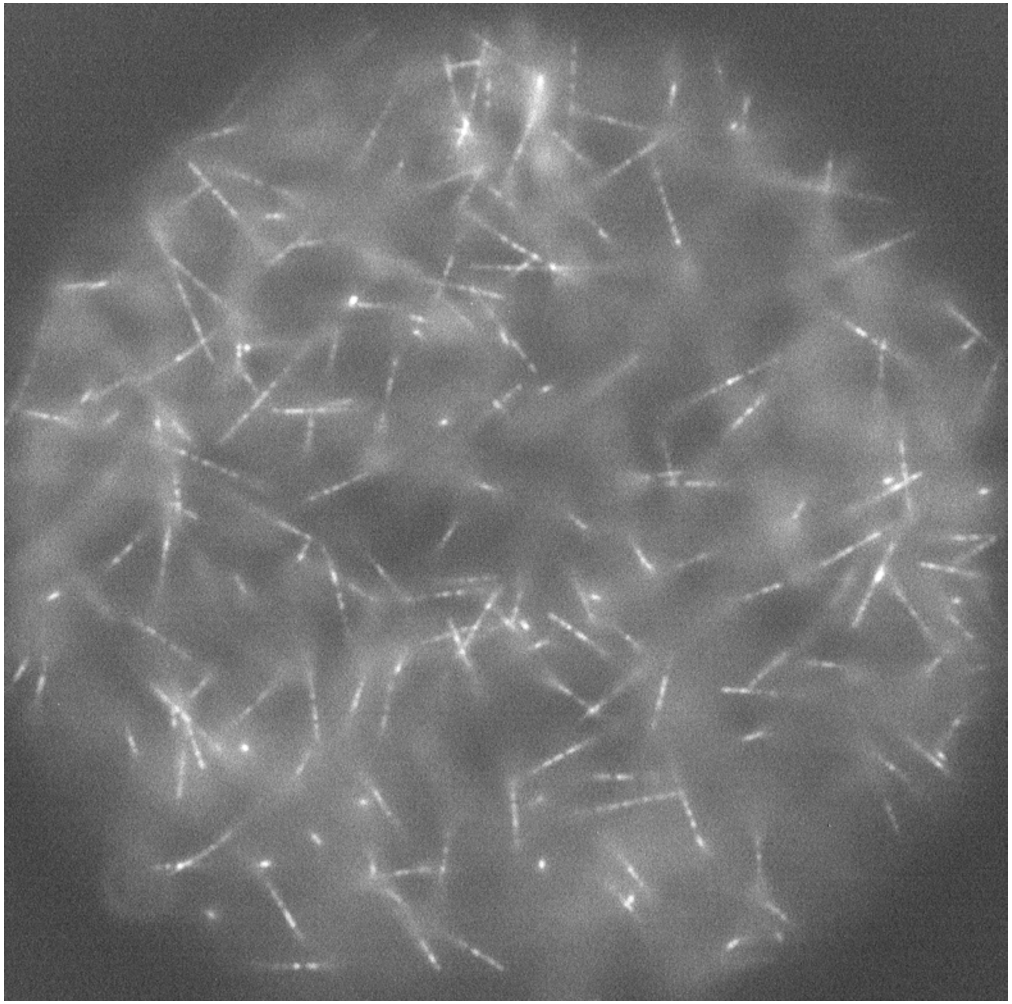


Alpha particle tracks (Am-241) in LiF FNTD



maximum intensity projection of a stack of 10 images (10 µm layer of a crystal)

Alpha particle tracks (Am-241) in LiF FNTD



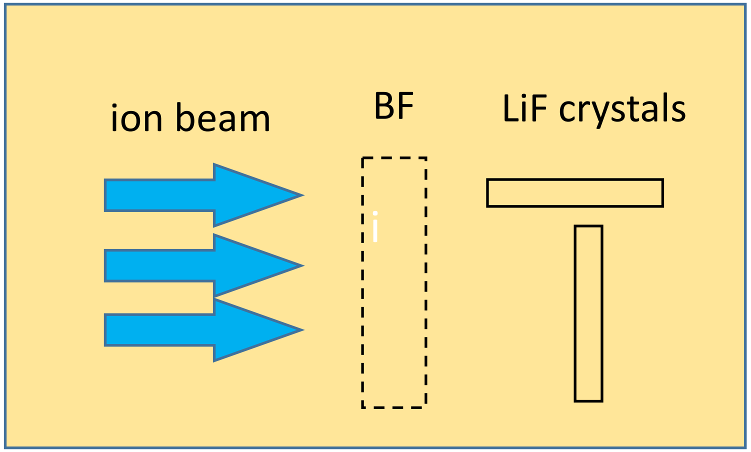
Short depth of focus (<1µm) enables vertical sectioning (”3D” images)

High-energy ions

HIMAC accelerator – several runs over year 2017-2019

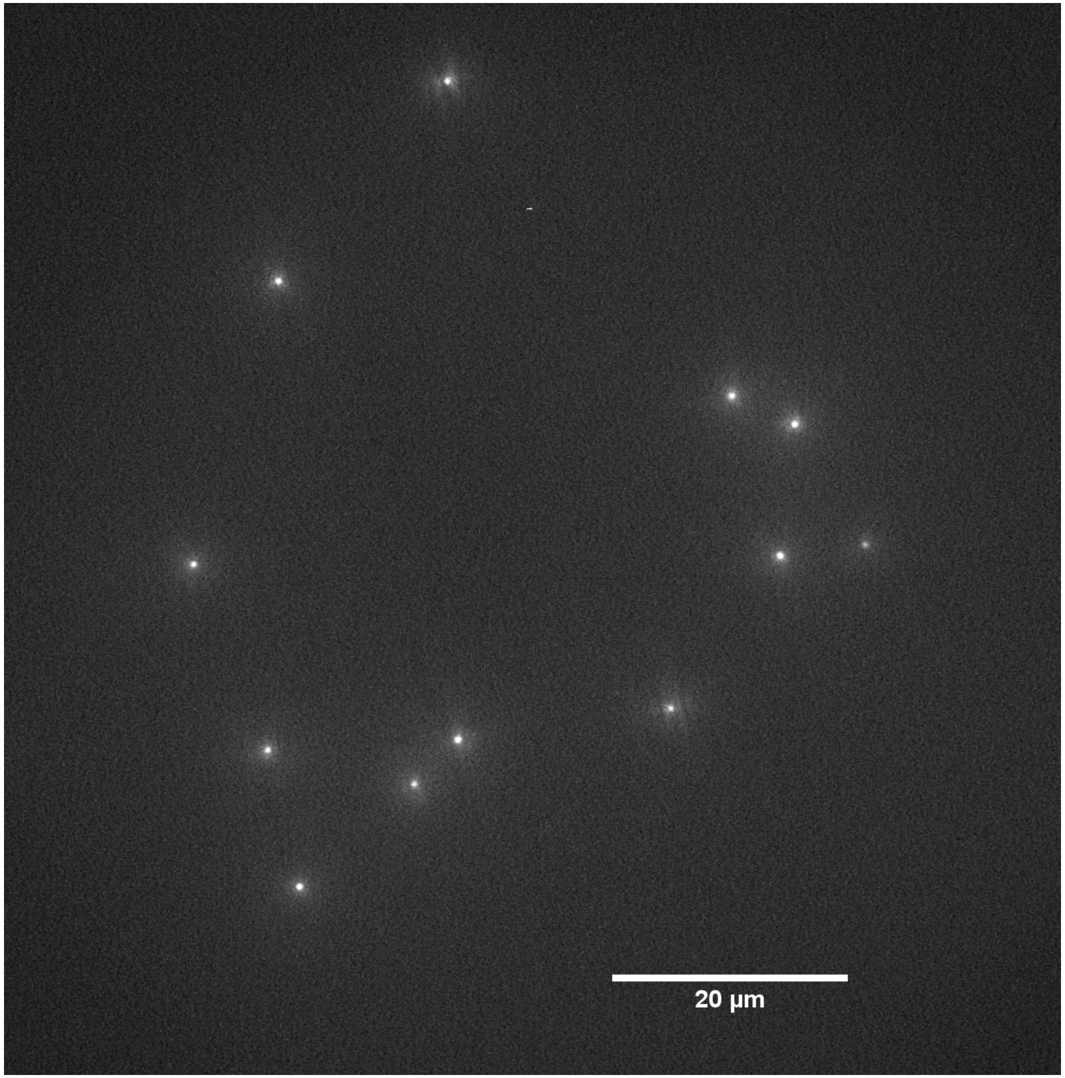
Carbon	290 MeV/nucleon
Carbon	400 MeV/nucleon
Neon	400 MeV/nucleon
Iron	500 MeV/nucleon
Helium	150/MeV/nucleon
Silicon	490 MeV/nucleon
Argon	500 MeV/nucleon

LiF crystals irradiated with two directions of the ion beam:



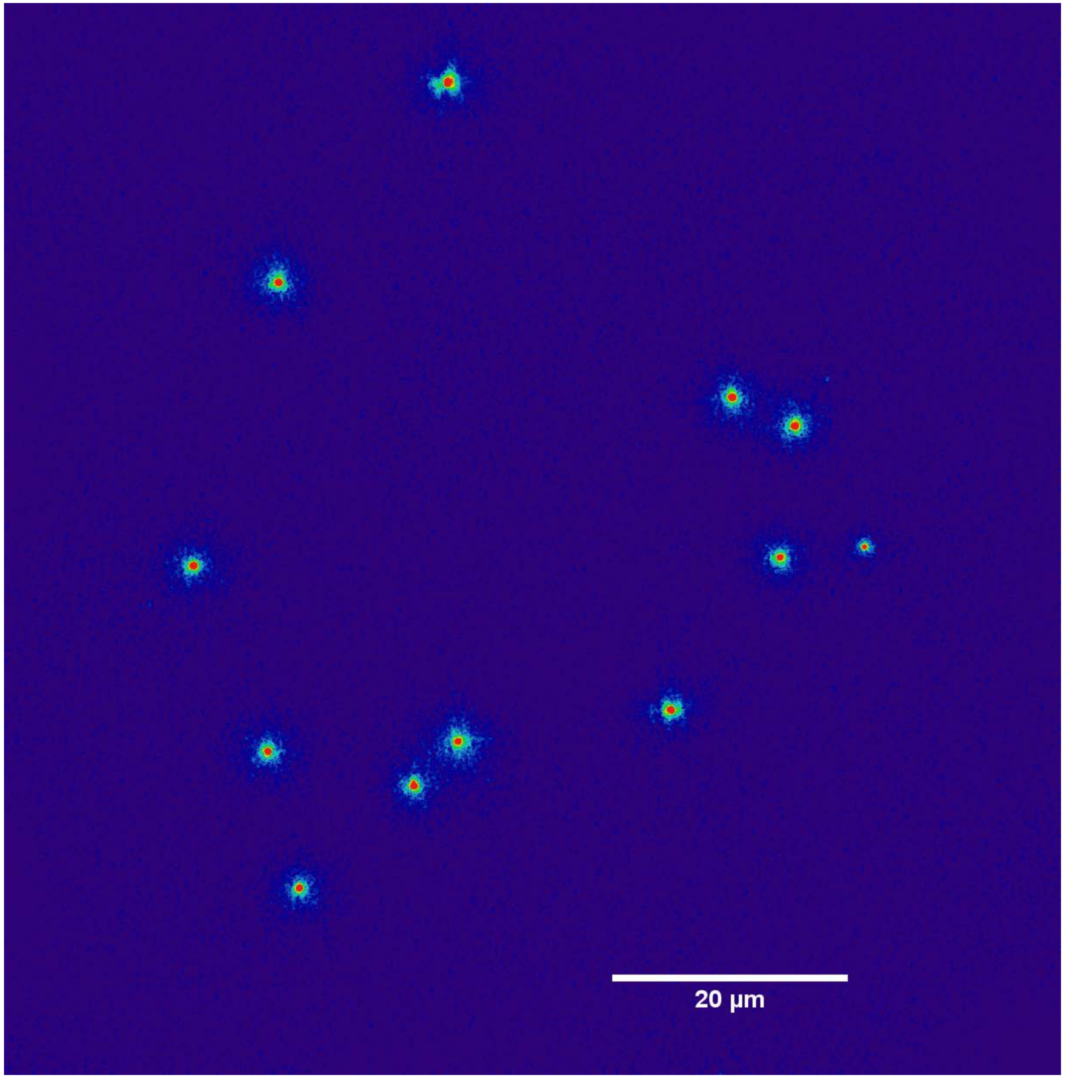
- perpendicularly to the surface
- (nearly) parallel to the surface

Iron ions, E=412 MeV/nucl, LET = 199 keV/µm

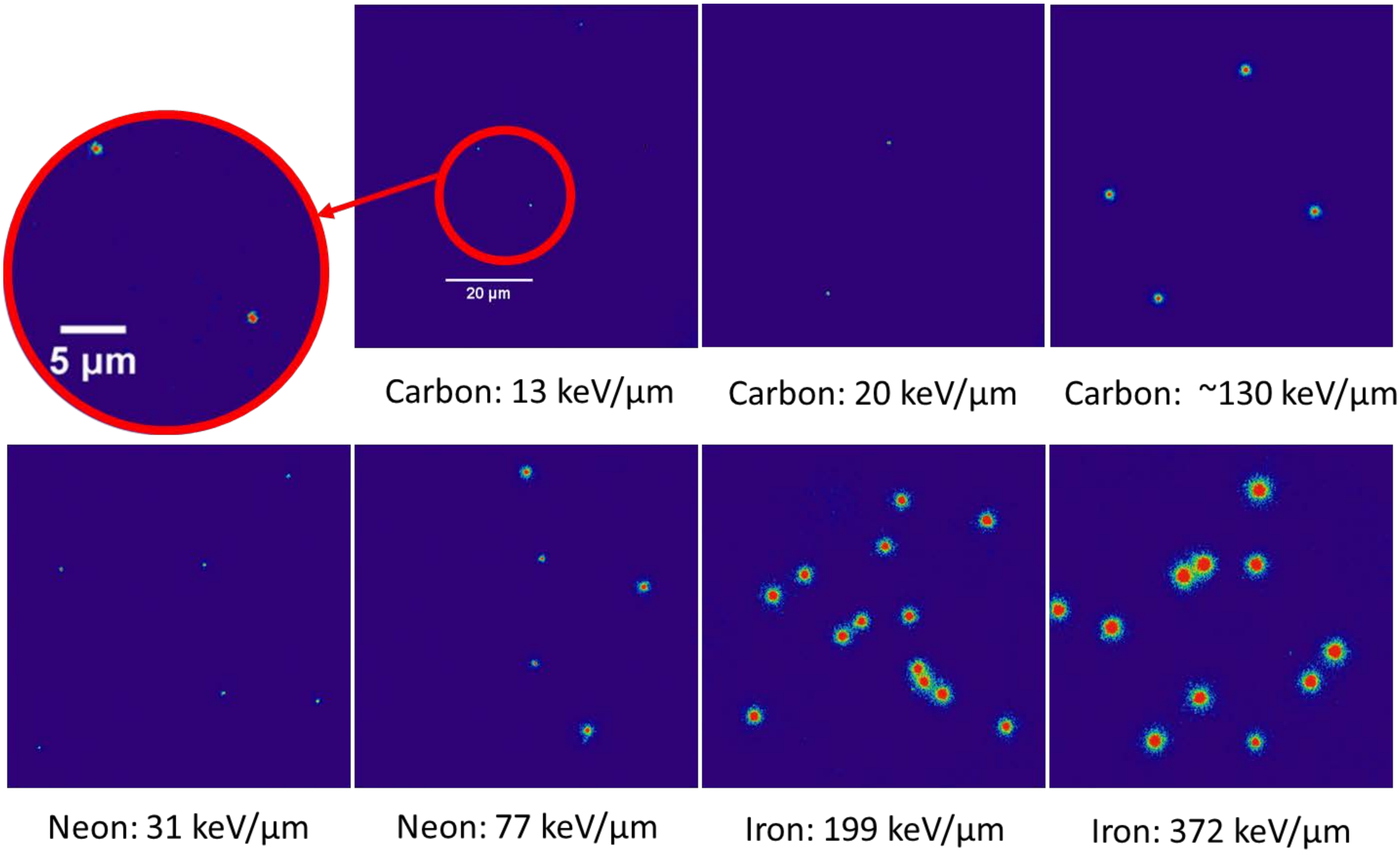


aquisition time 10s, depth in crystal 18µm, original image

Iron ions, E=412 MeV/nucl, LET = 199 keV/µm



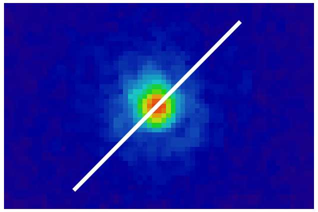
aquisition time 10s, depth in crystal 18µm; image after background subtracting

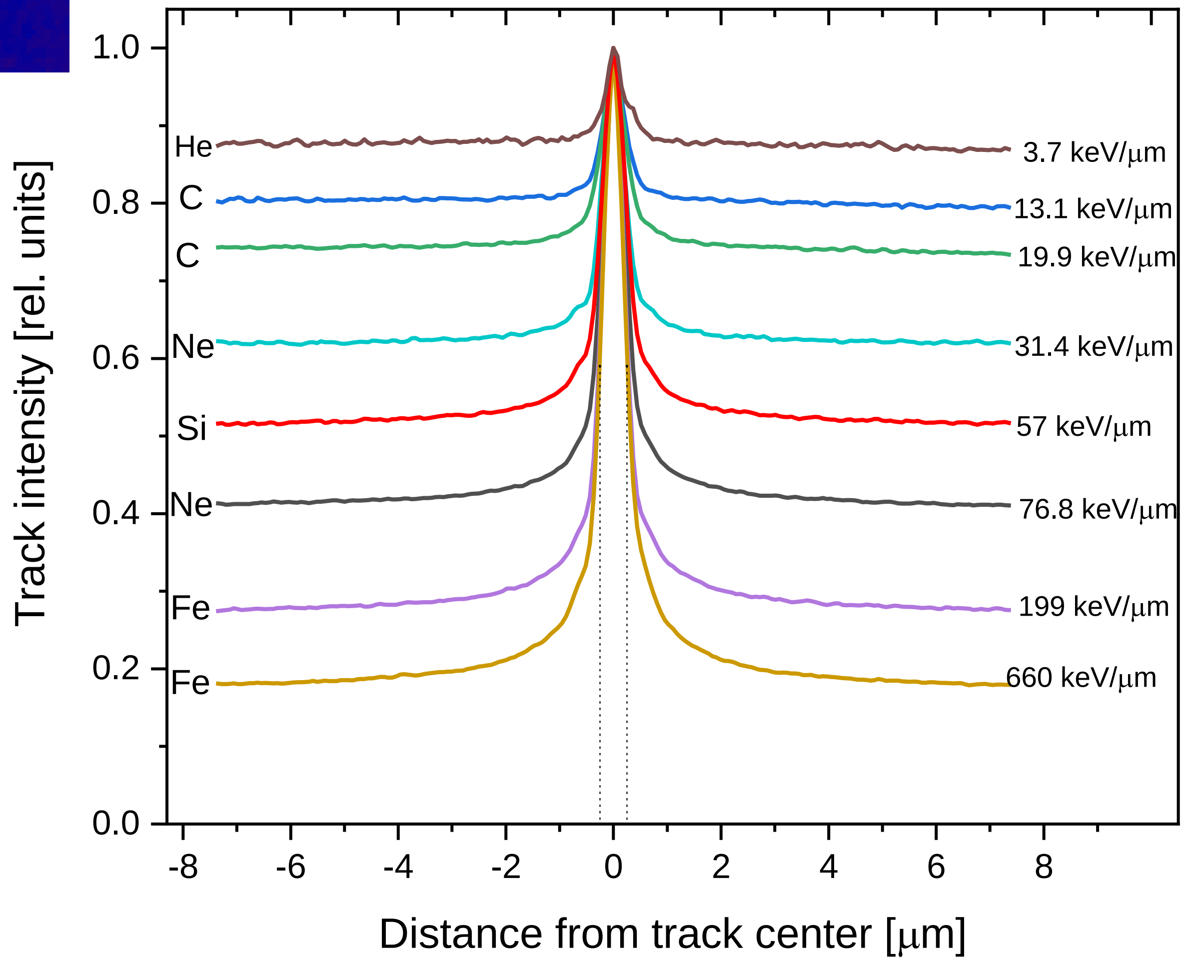

5 µm
20 µm
Carbon: 13 keV/µm
Carbon: 20 keV/µm
Carbon: ~130 keV/µm
Neon: 31 keV/µm
Neon: 77 keV/µm
Iron: 199 keV/µm
Iron: 372 keV/µm

(LET in water)

Track profiles

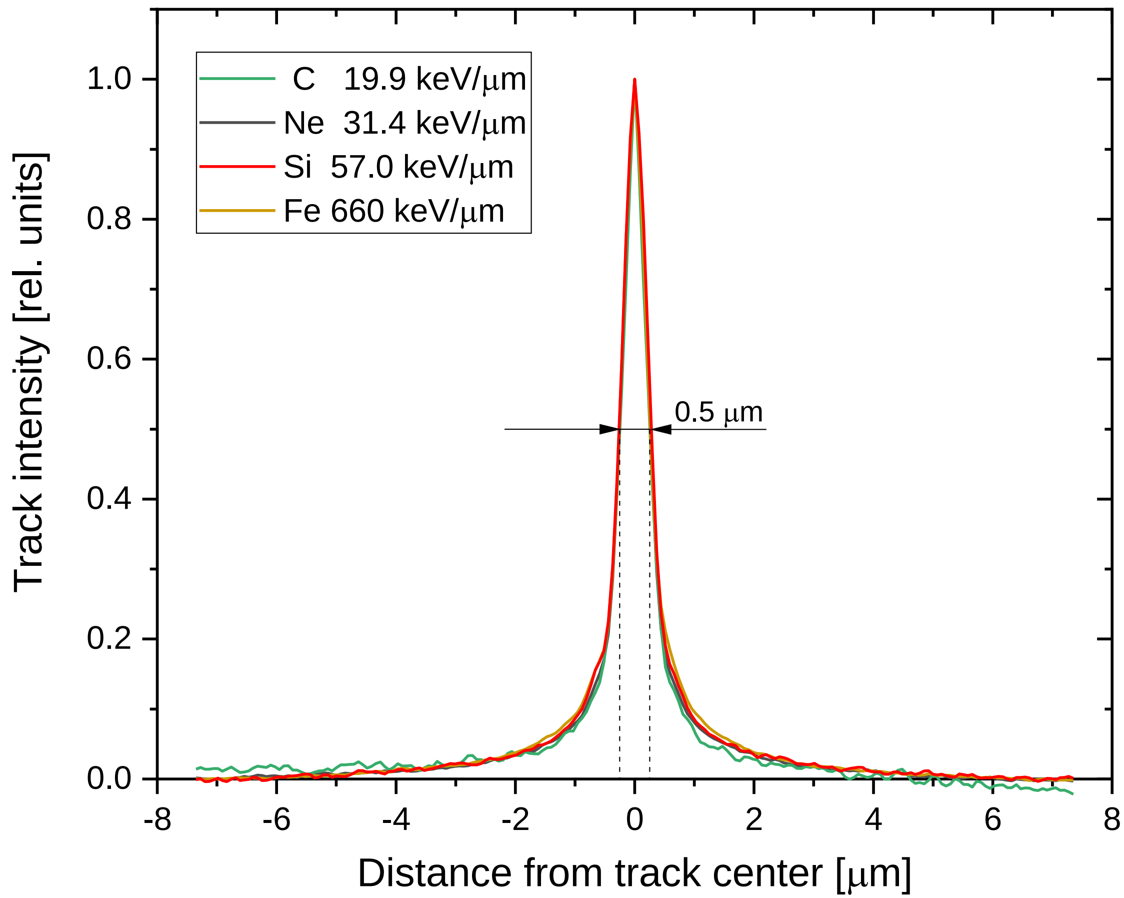
(intensity along diameter lines)





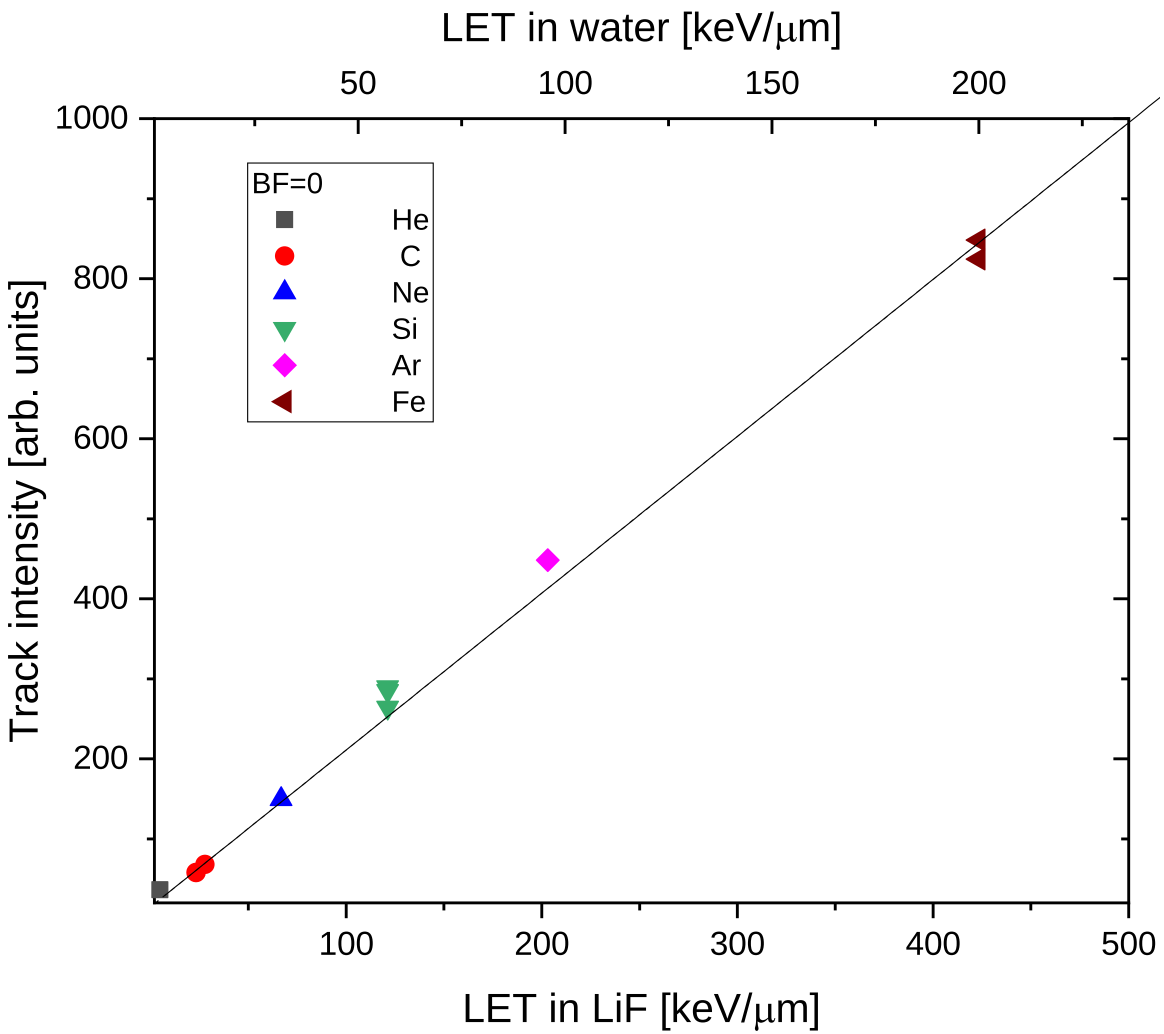
Track profiles

normalized after background subtraction

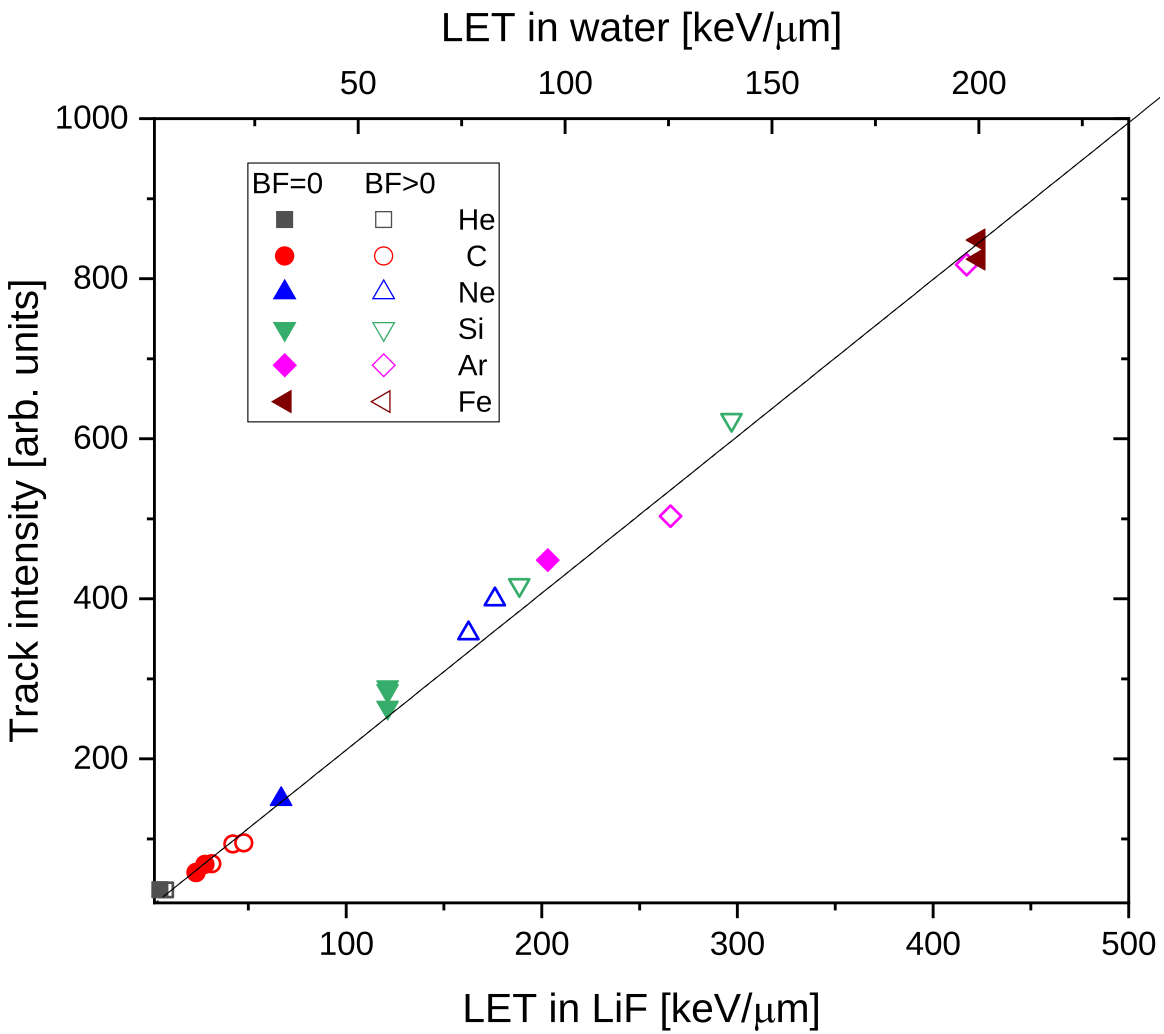


- Track shape identical for all ions (differences only in intensity)
- FWHM corresponds with the theoretical resolution limit of the microscope: c.a. 400 nm

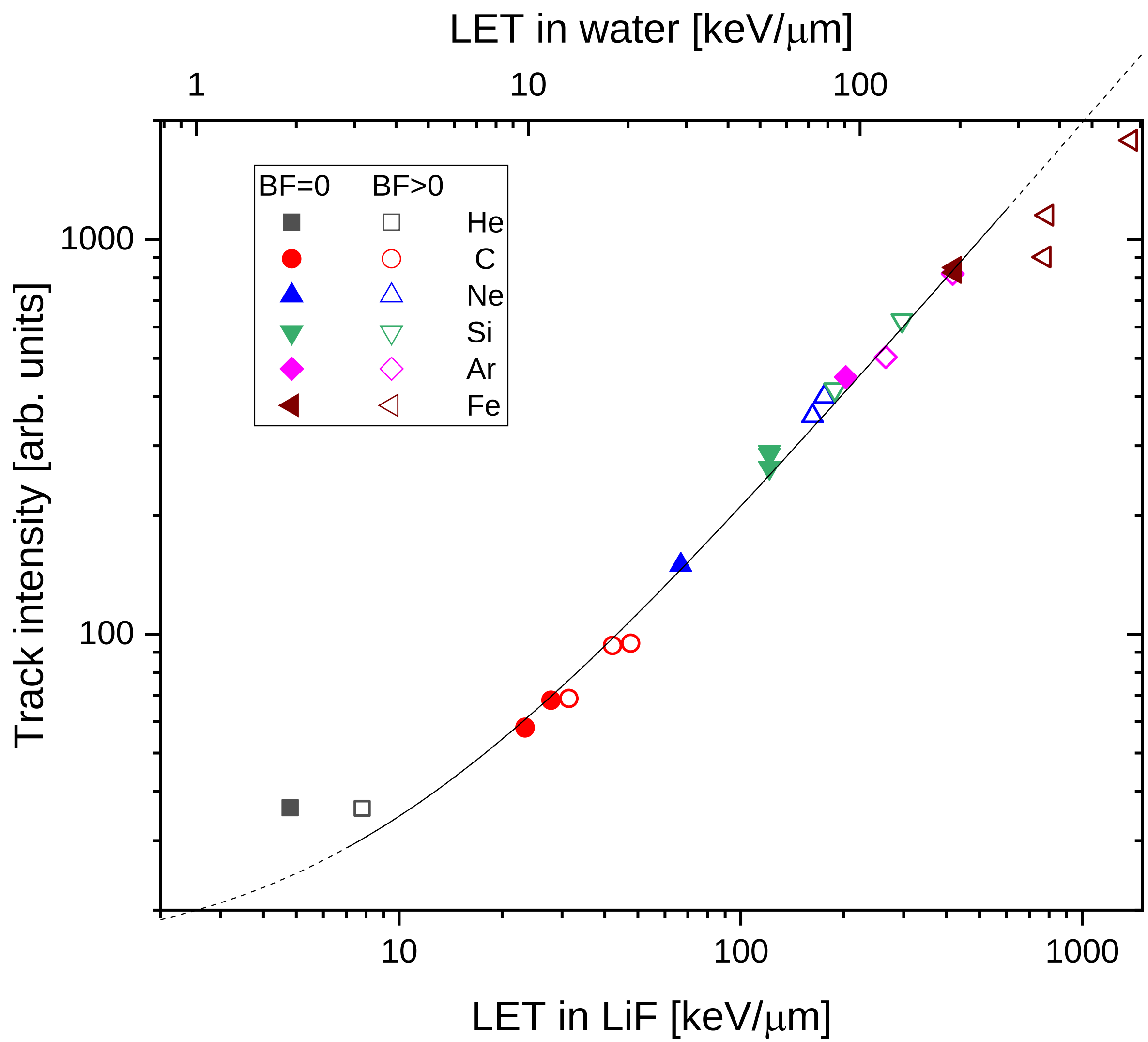
Track intensity vs. LET



Track intensity vs. LET



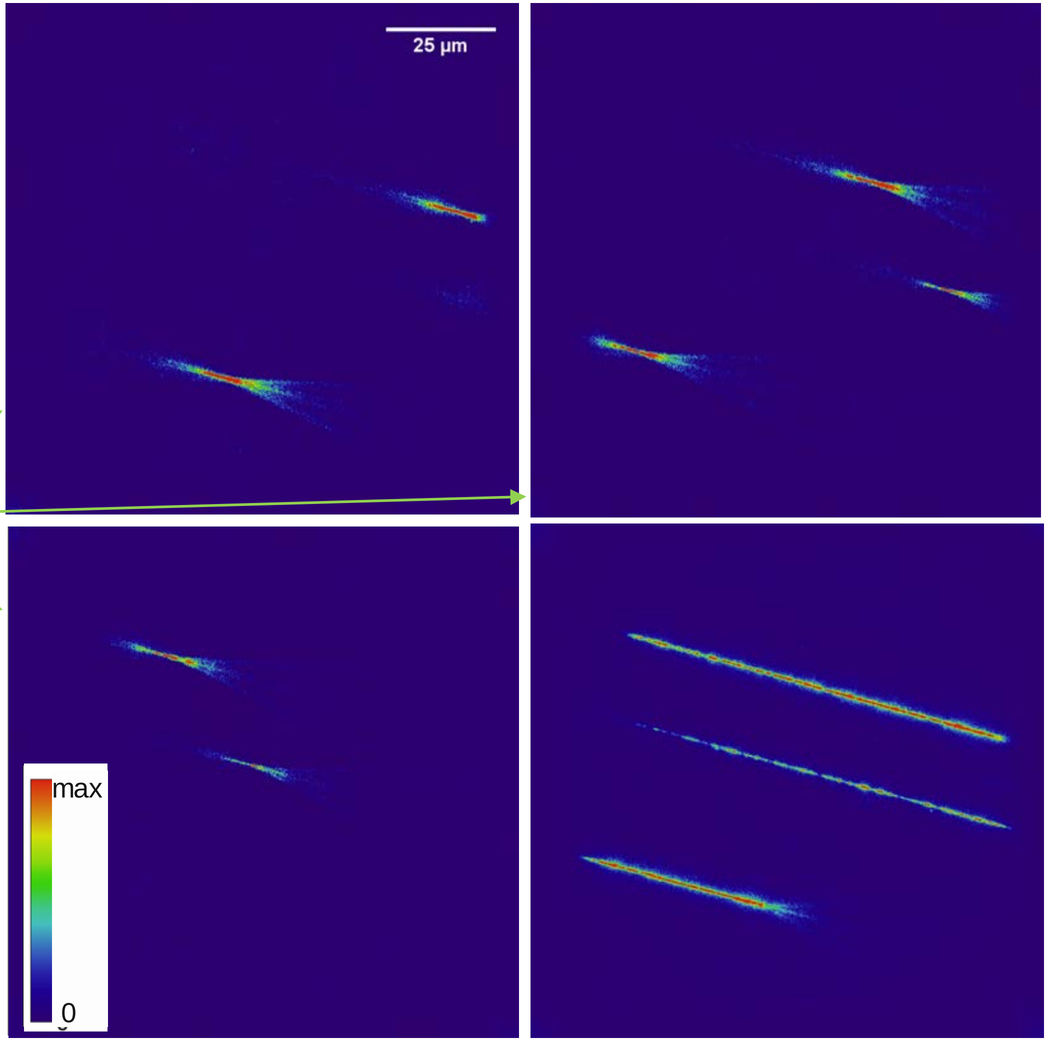
Track intensity vs. LET



Tracks parallel to crystal surface

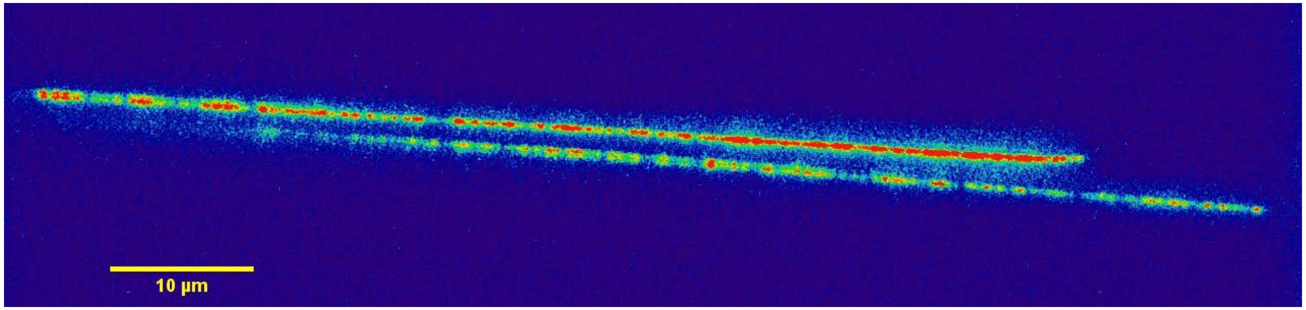
Iron ions, E=145 MeV/n, LET = 370 keV/µm

single frames of a stack registered at various depths

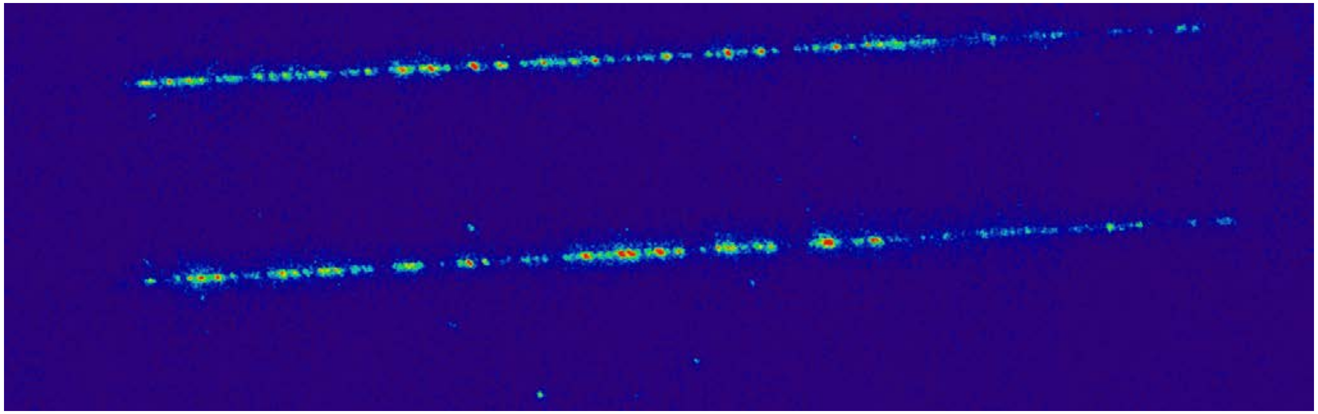


maximum intensity projection of the whole stack

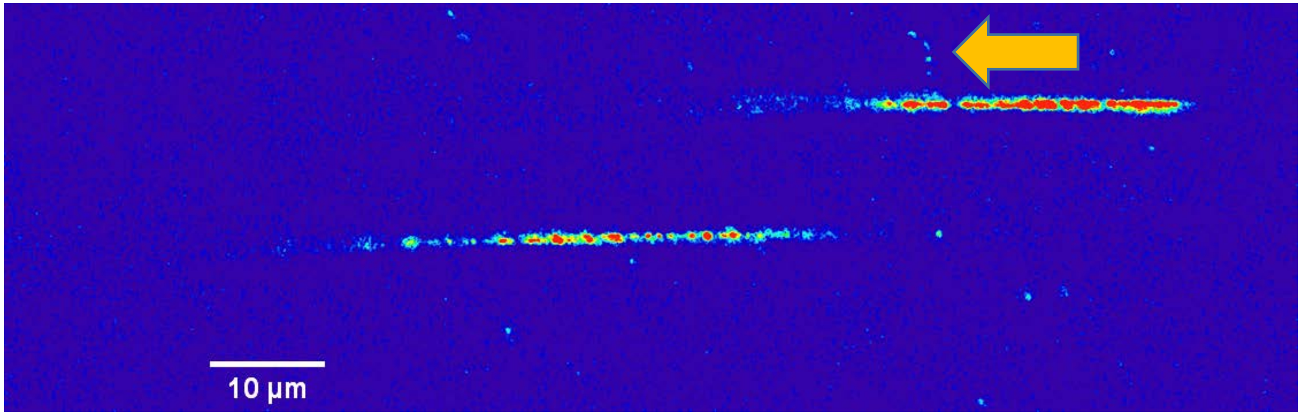
Carbon ions
stopping in crystal



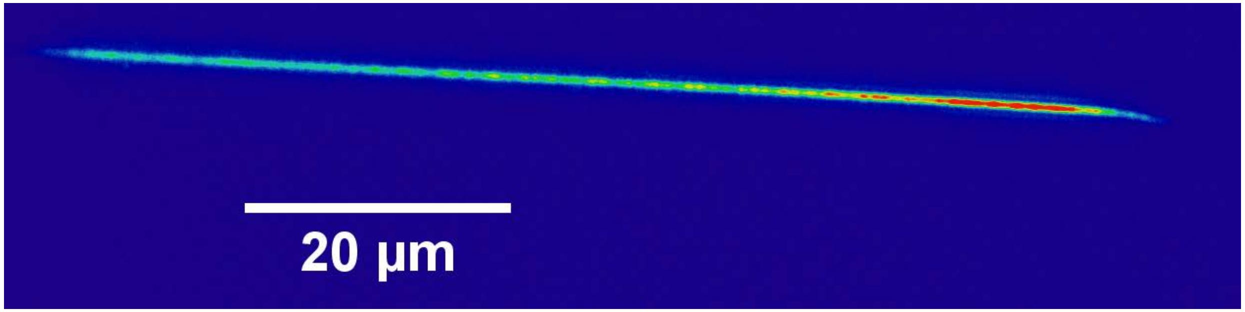
Si ions,
E= 442 MeV/n
(at entering crystal)

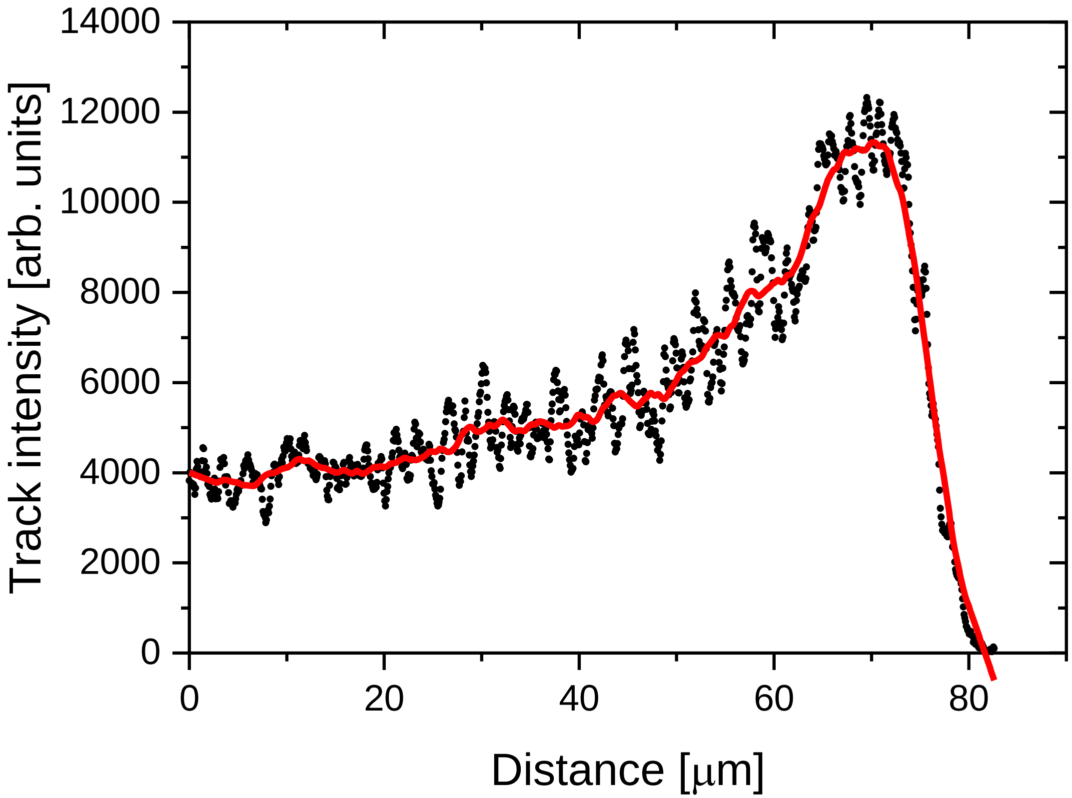


Ne ions
E =94 MeV/n
(at entering crystal)



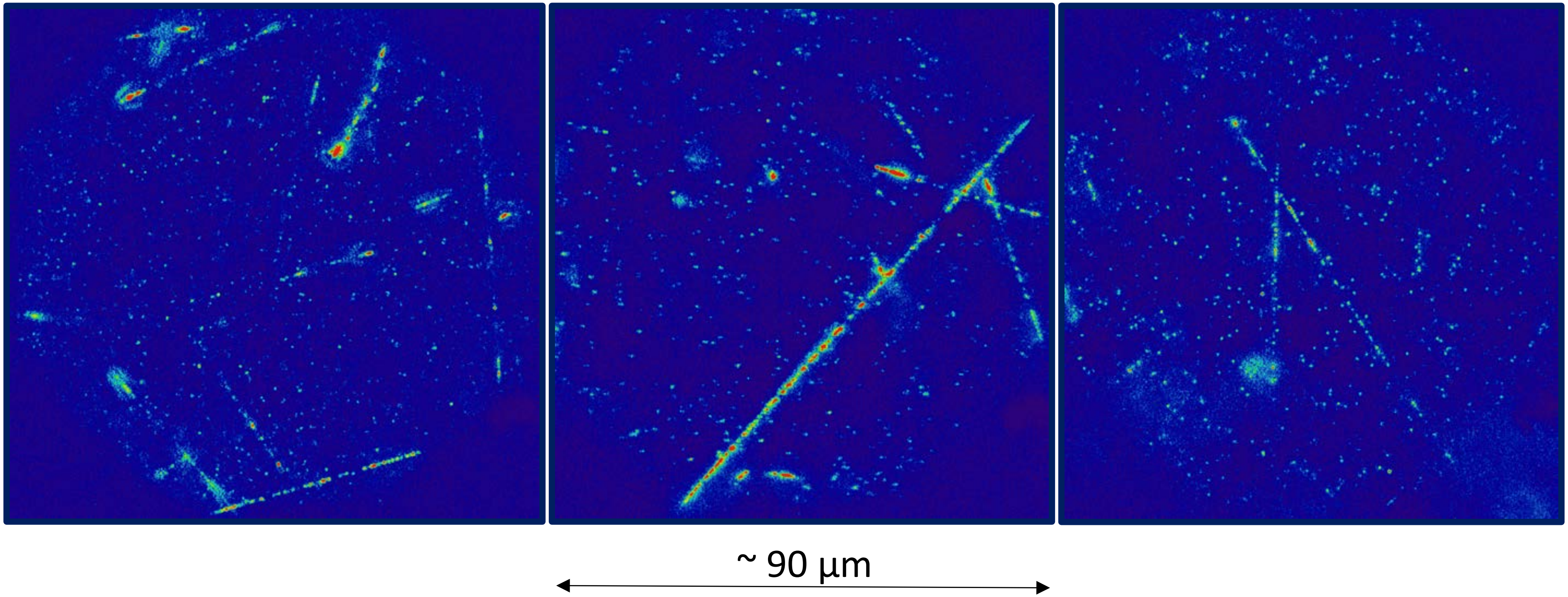
Iron ion stopping in the crystal





Cosmic radiation

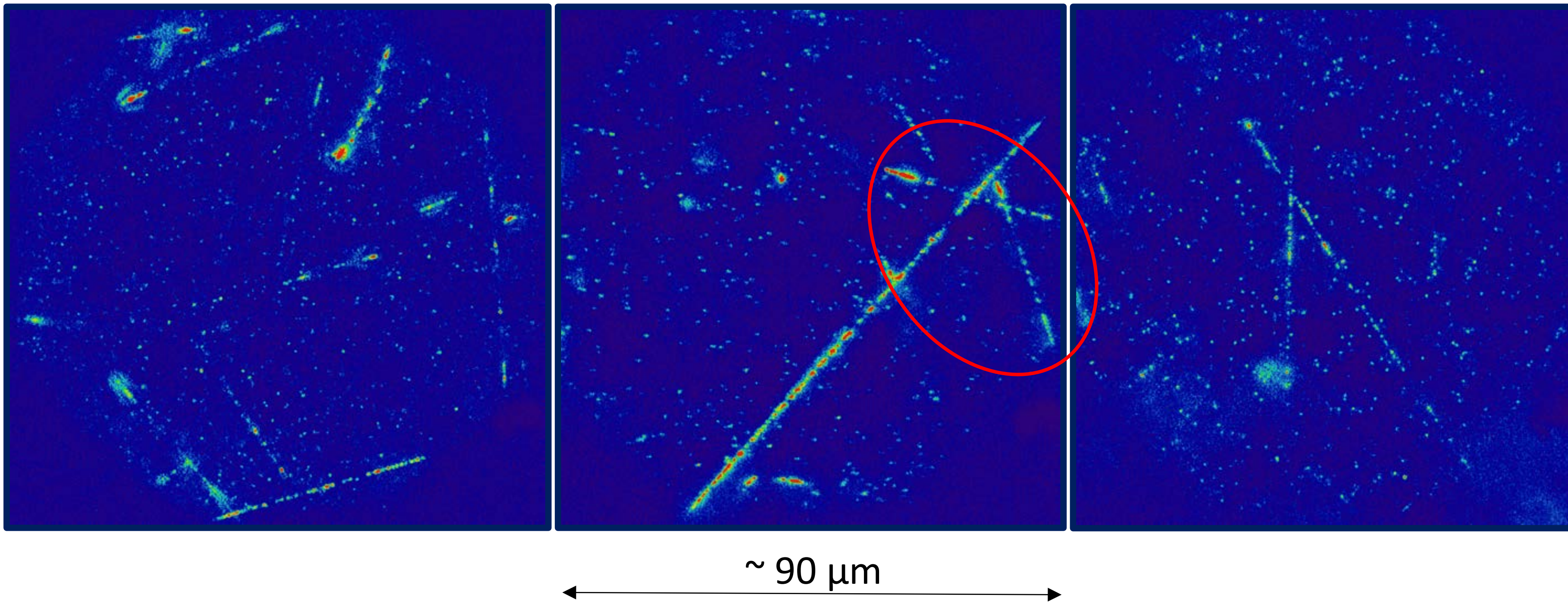
DOSIS 3D (#13) - 6 months on ISS



quantitative analysis not yet available…

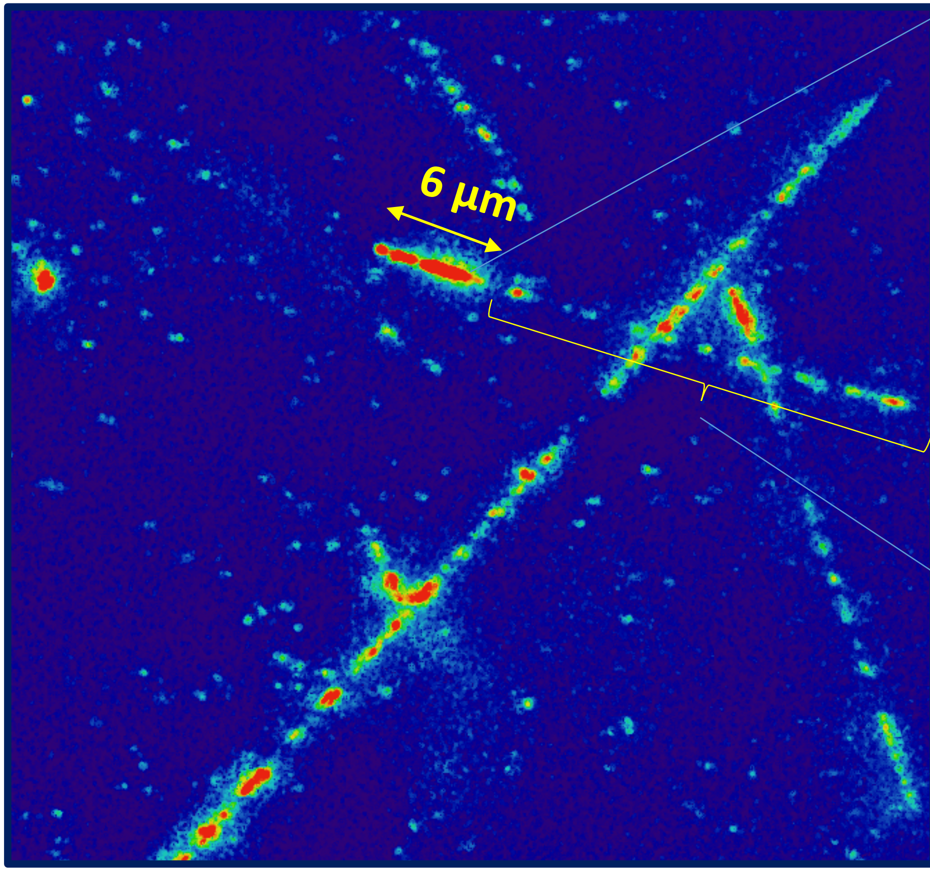
Cosmic radiation

DOSIS 3D (#13) - 6 months on ISS



quantitative analysis not yet available…

Neutrons: $^{6}Li(n,\alpha)^{3}H$



α particle ^{4}He, E_{α}=2.05 MeV
range in LiF 6.05 µm

Triton ^{3}H, E_{T}=2.73 MeV
range in LiF 33.5 µm

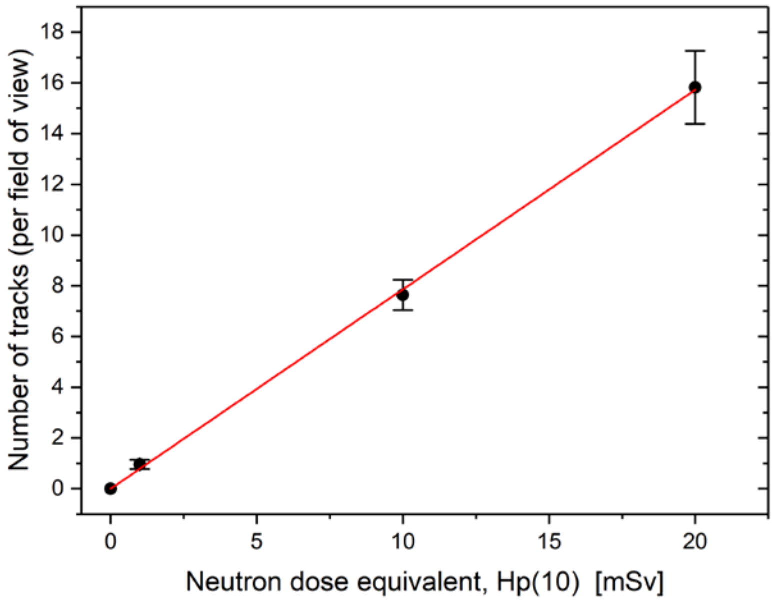
Personal neutron dosimetry: *albedo* technique

(exposures on a PMMA slab pantom)

- for Pu-Be source: **1 track/mSv/field_of_view** (∅ 90µm)
- **no tracks for zero dose!**

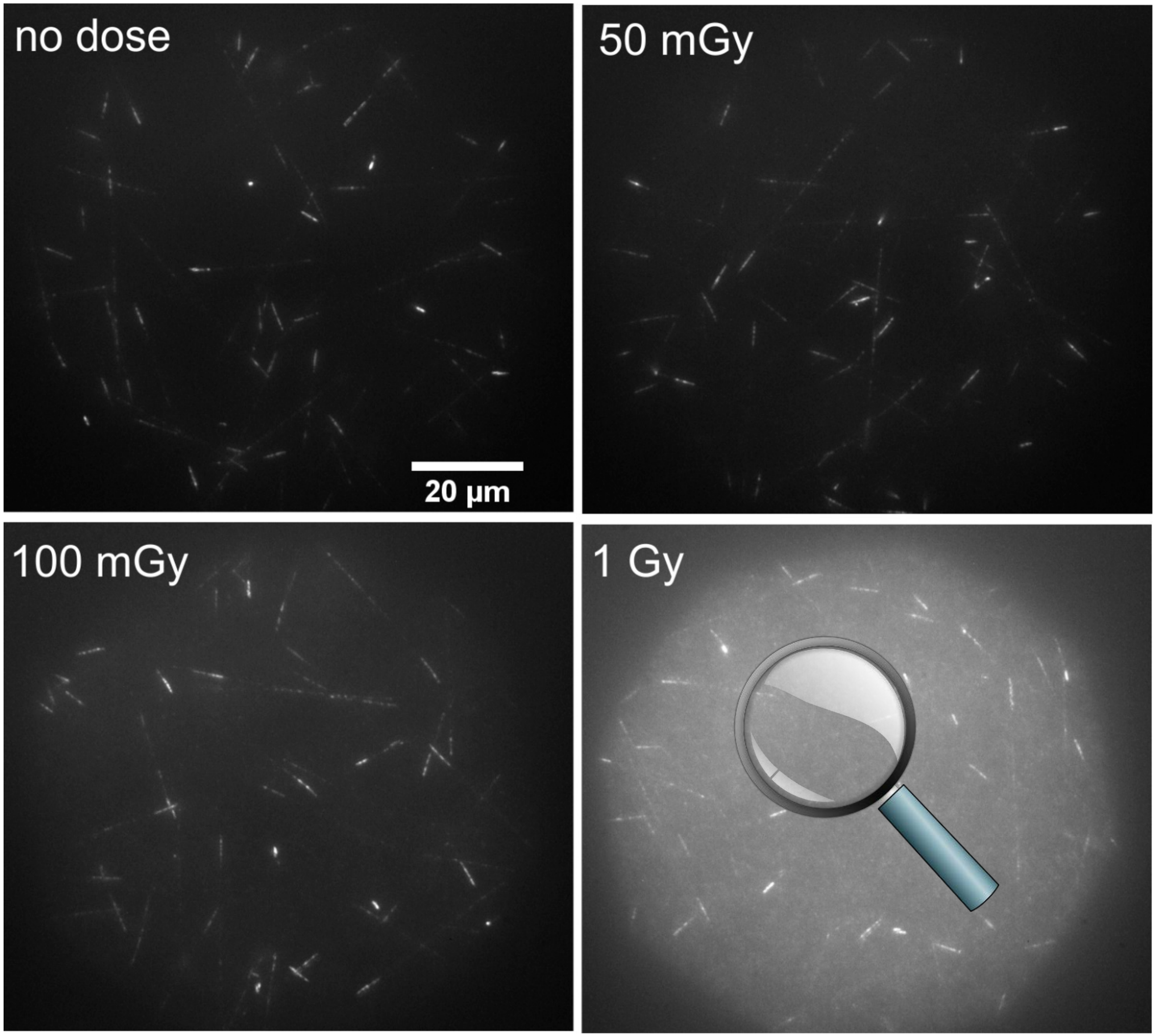
detection limit may be very low - depending on the analyzed volume of a crystal

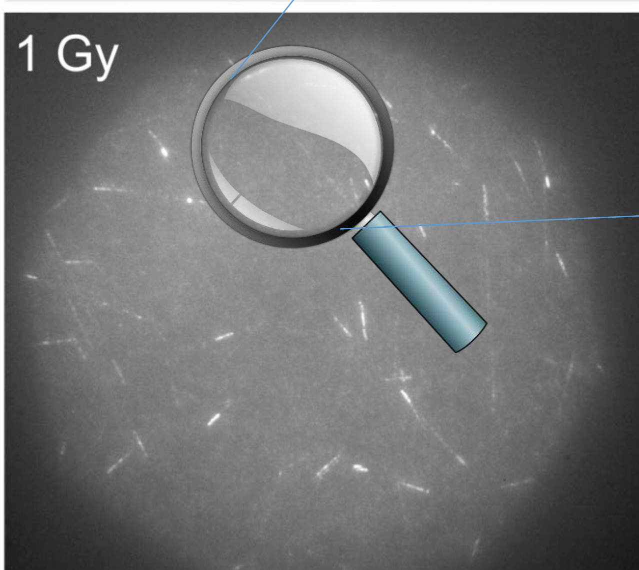
for ^{6}Li enriched LiF efficiency will be still much increased
(^{6}LiF crystal recently grown – work under way)

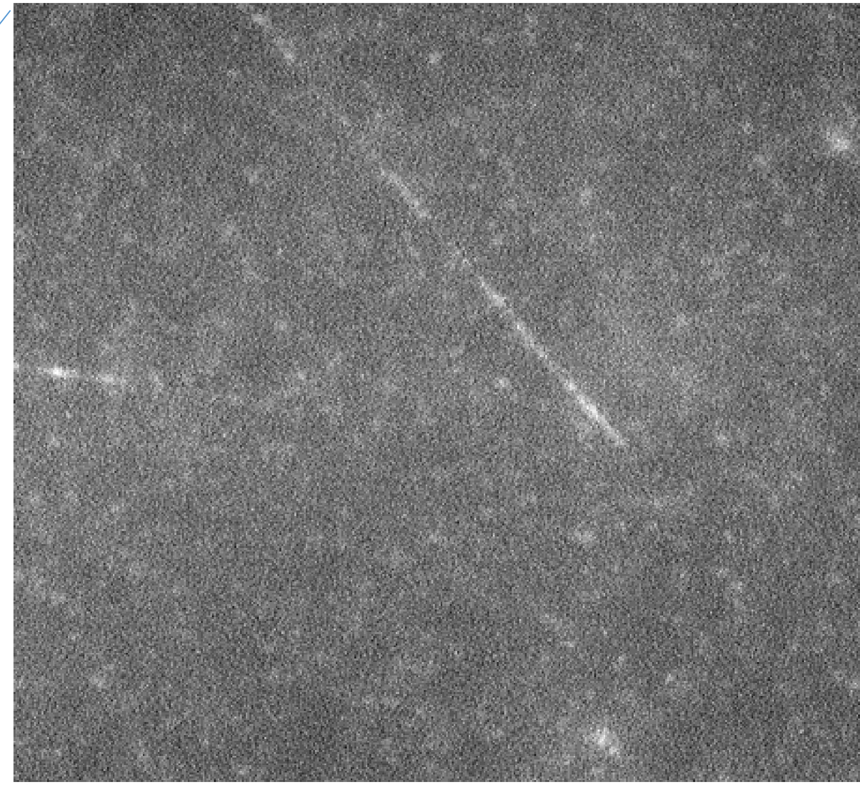


P. Bilski, B. Marczewska, M. Kłosowski, W. Gieszczyk, M. Naruszewicz, *Detection of neutrons with LiF fluorescent nuclear track detectors* Radiat. Meas. 116 (2018) 35-39

Mixed field: neutrons + gamma radiation

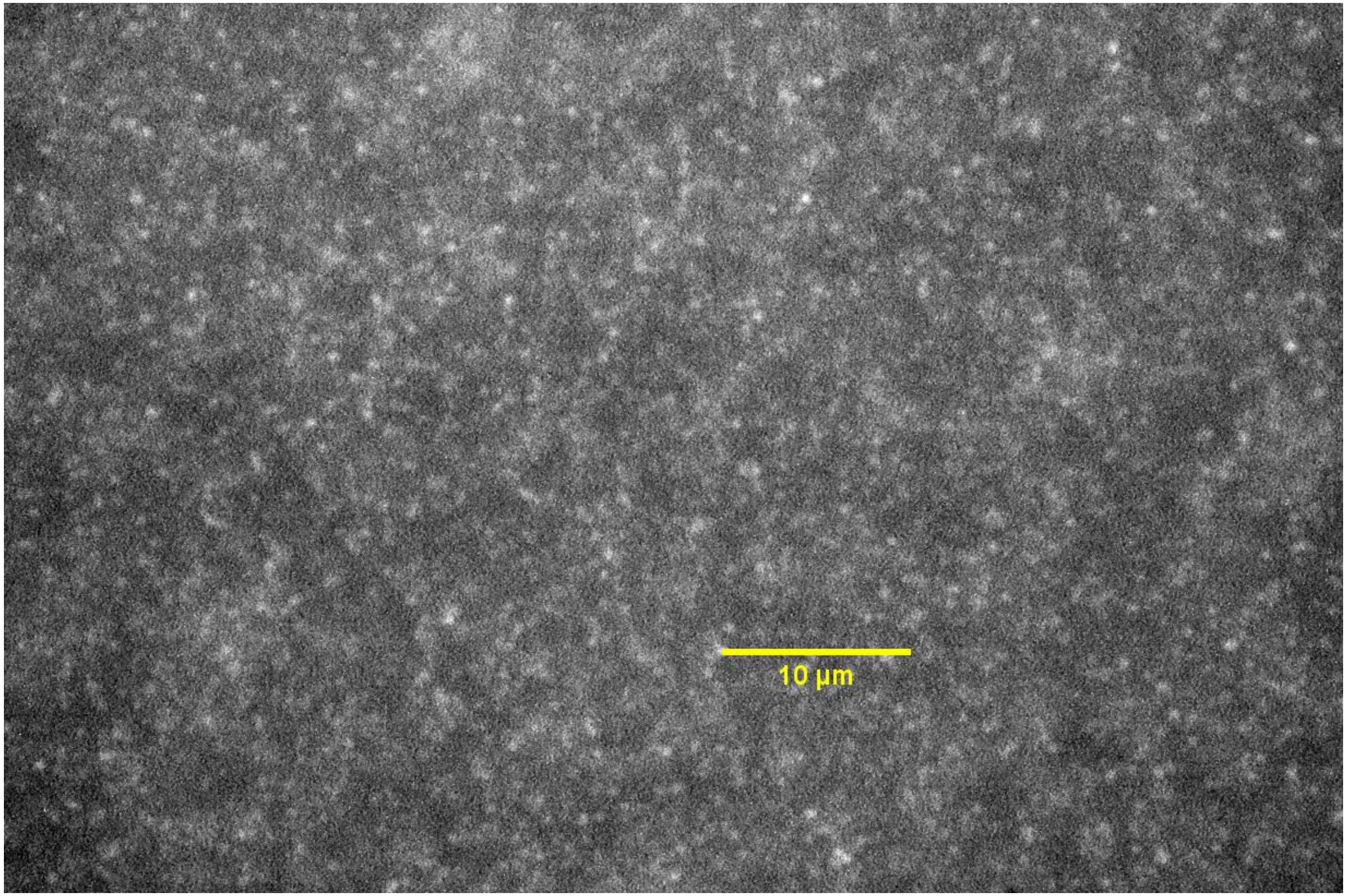






Gamma-ray induced background is not uniform, but consists of small spots (<1 µm)!

X-rays



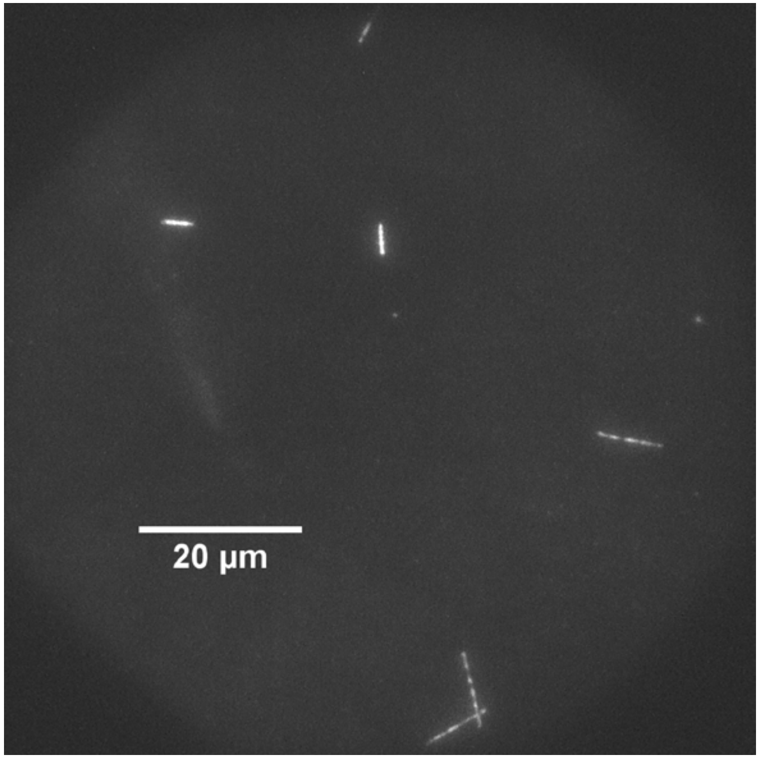
X-rays, **9 keV**, 150 mGy

secondary electrons → possibly we see the end of the particle path

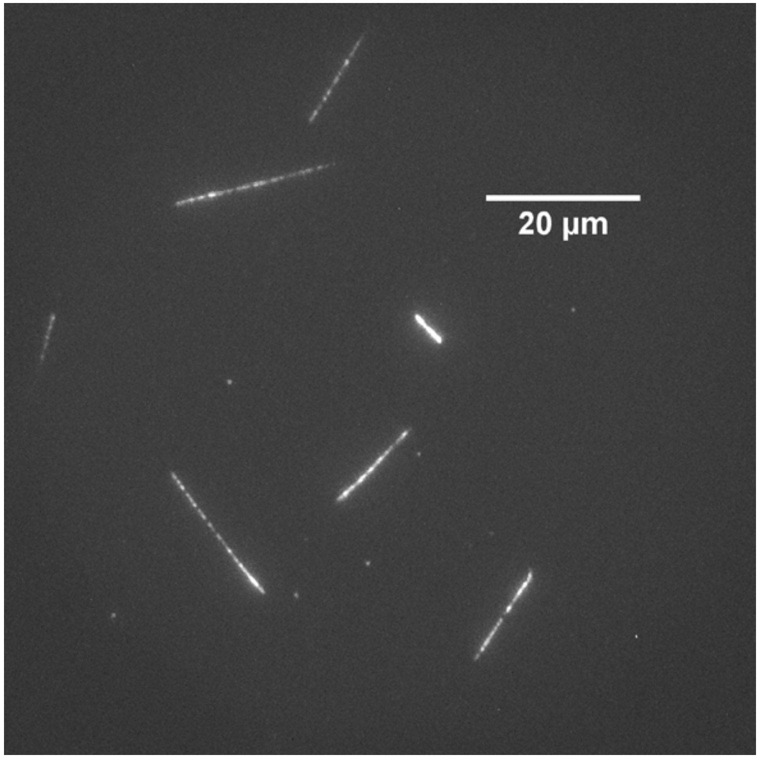
Range-based spectrometry
for stopping particles

tracks of alpha particles with different energy

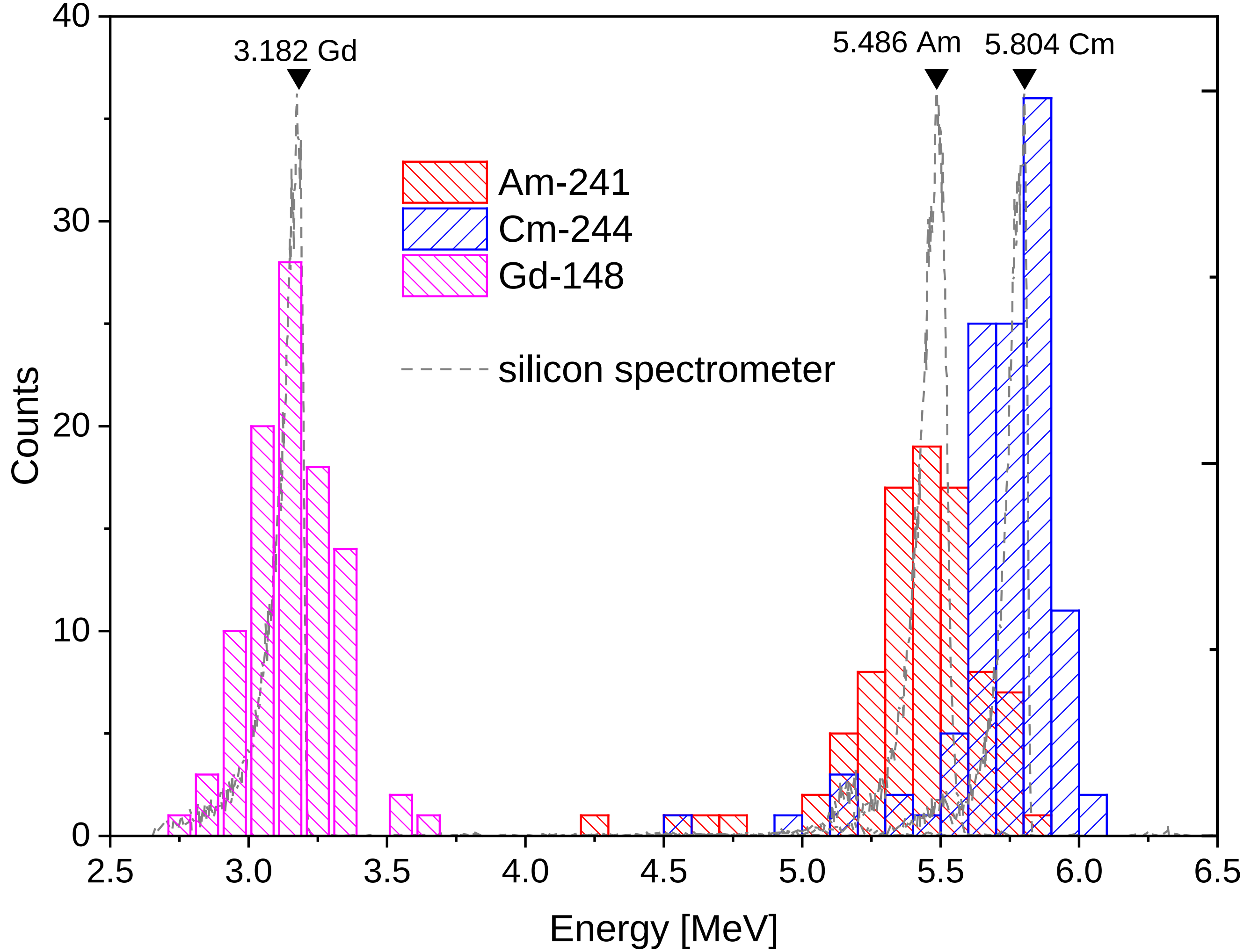
Gd-148, E=3.182 MeV



Cm-144, E=5.804 MeV



Range-based spectrometry



Concluding remarks

- The fluorescent tracks were observed in LiF crystals for variety of particles (even electrons), including the cosmic radiation field at ISS
- Track intensity (brightness) was found to be directly related to LET, so LET measurements seem to be possible, but much more work is still needed to obtain reliable quantitative results
- The most straightforward application of LiF FNTDs is neutron dosimetry using products of $^{6}Li(n,\alpha)$ reaction, which may allow measurements of very low doses even in high gamma background

Thank you for attention!

PUBLICATIONS:

- *Fluorescent imaging of heavy charged particle tracks with LiF single crystals,* J. Lumin. 213 (2019) 82-87;
- *Luminescent properties of LiF crystals for fluorescent imaging of nuclear particles tracks,* Opt. Mat. 90 (2019) 1-6;
- *Fluorescent detection of single tracks of alpha particles using lithium fluoride crystals*, Nucl. Instr. Meth. B 392 (2017) 41–45;
- *Lithium fluoride crystals as Fluorescent Nuclear Track Detectors,* Radiat. Prot. Dosim. 178 (2018) 337-340;
- *Detection of neutrons with LiF fluorescent nuclear track detectors, Radiat* Meas. 116 (2018) 35-39.