TU WIEN
TECHNISCHE UNIVERSITÄT WIEN
Vienna | Austria

ATOMINSTITUT

# Neutron irradiation experiments on HTS

**Raphael Unterrainer[1] , Davide Gambino[2], Florian Semper[1], Alexander Bodenseher[1], Danielle Torsello[3,4], Francesco Laviano[3,4], David X. Fischer[5]**
**Michael Eisterer[1]**

[1]Atominstitut, TU Wien, Vienna, Austria
[2]IFM, Linköping University, Linköping, Sweden
[3]DiSAT, Politecnico di Torino, Torino, Piemonte, Italy
[4]INFN, Sezione di Torino, Torino, Piemonte, Italy
[5]PSFC, MIT, Cambridge, Massachusetts, United States

**RADSUM, CERN, January 16th 2025**

# Collaborations and Funding

EUROfusion

This work has been carried out within the framework of the EUROfusion Consortium, funded by the European Union via the Euratom Research and Training Programme (Grant Agreement No 101052200 — EUROfusion). Views and opinions expressed are however those of the author(s) only and do not necessarily reflect those of the European Union or the European Commission. Neither the European Union nor the European Commission can be held responsible for them.

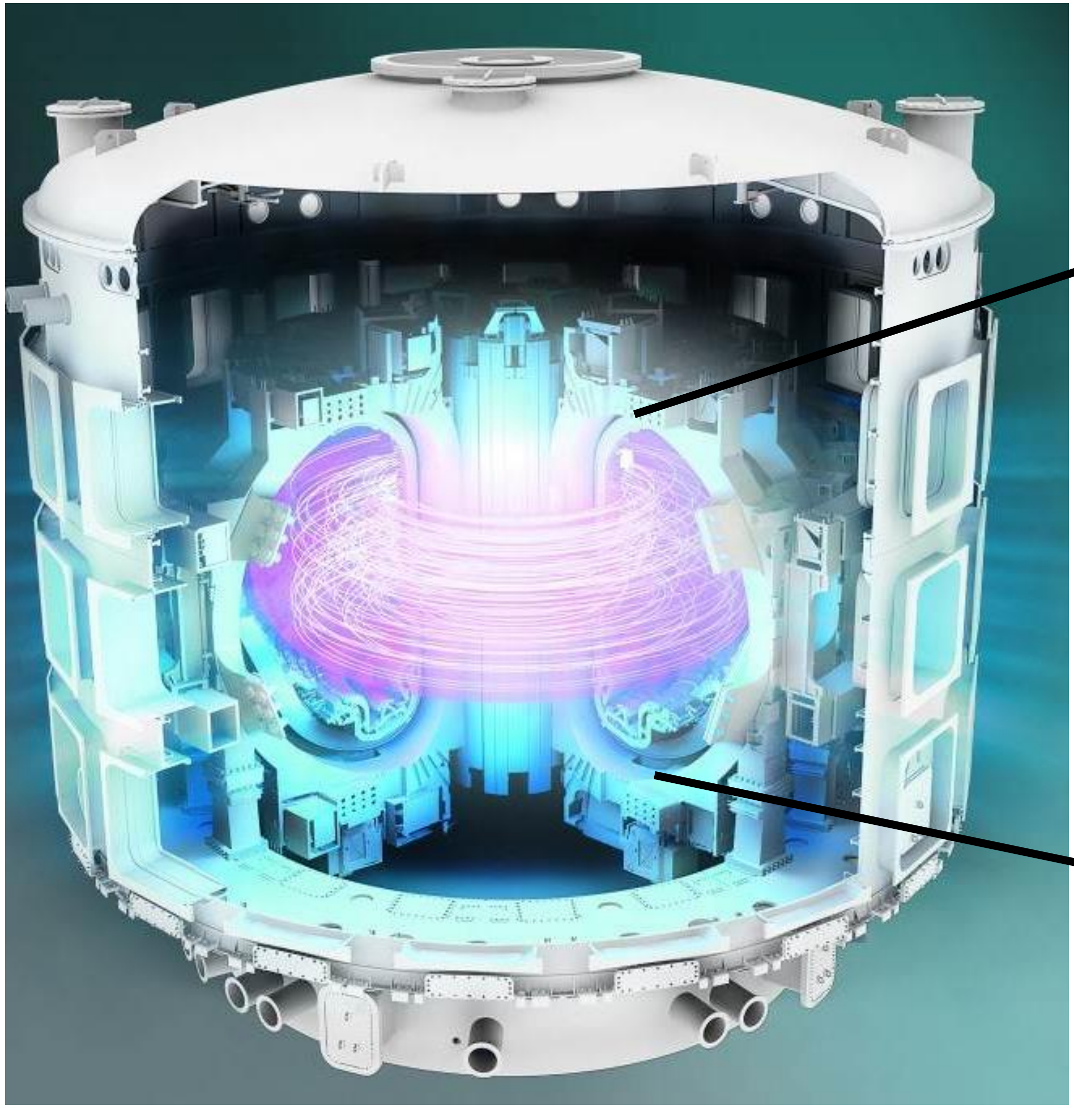
Funded by the European Union

Politecnico di Torino
1859

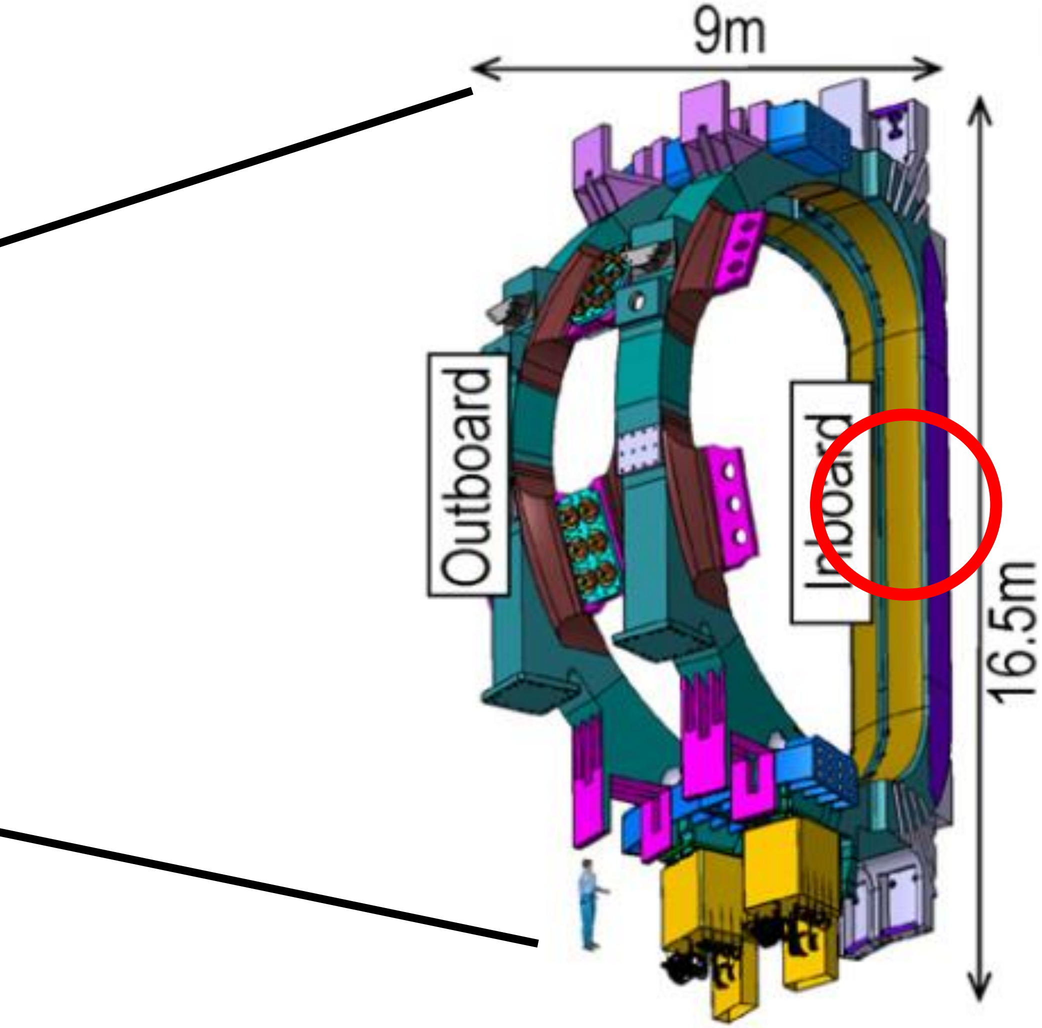
LINKÖPING UNIVERSITY

PSFC

# Irradiation Damage

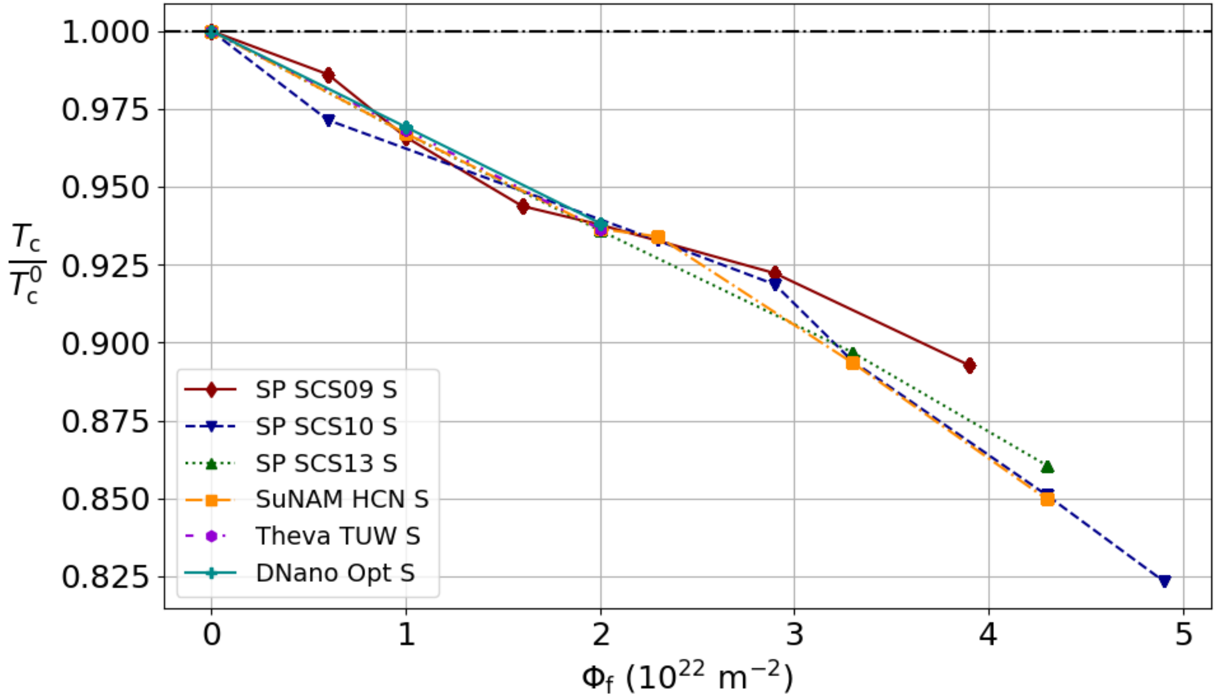


© ITER Organization, http://www.iter.org/



https://doi.org/10.1016/j.fusengdes.2008.12.105

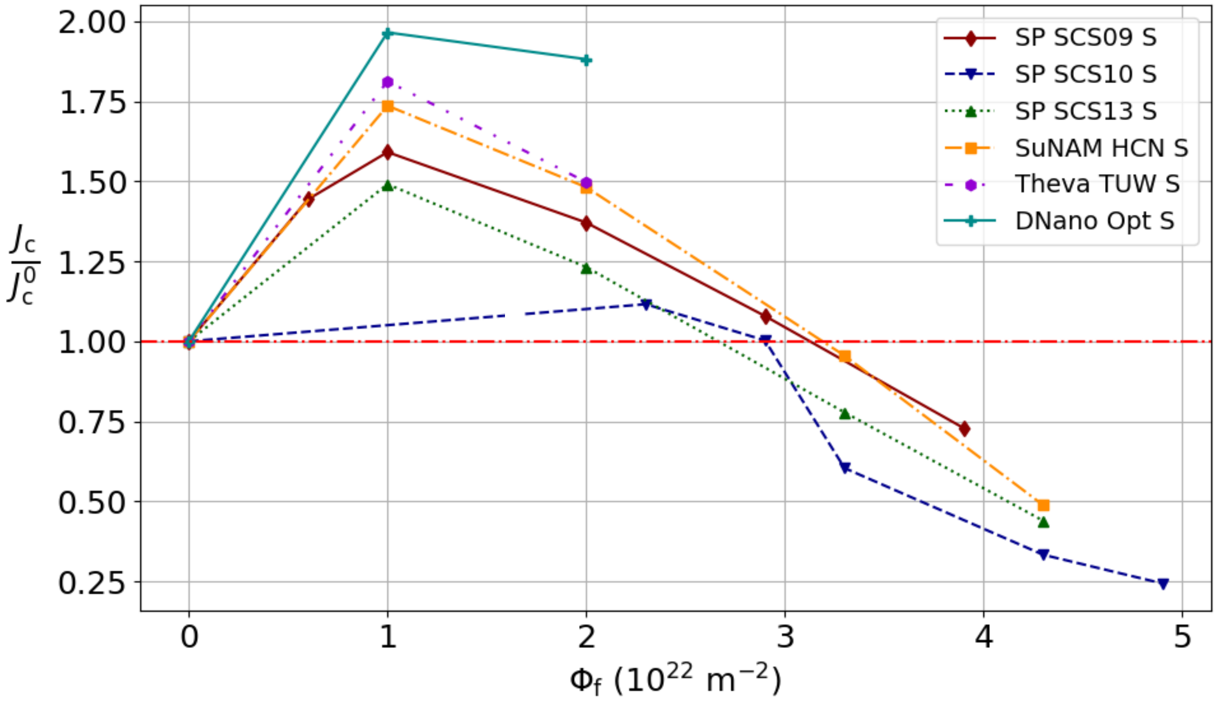
# Irradiation Damage



- Compact fusion devices – high fields for confinement
  - Currently REBCO coated conductors most promising
  - High quality, long lengths (800+ m)
  - Change of properties under irradiation conditions “well” known
  - Monotonic decrease of $T_c$

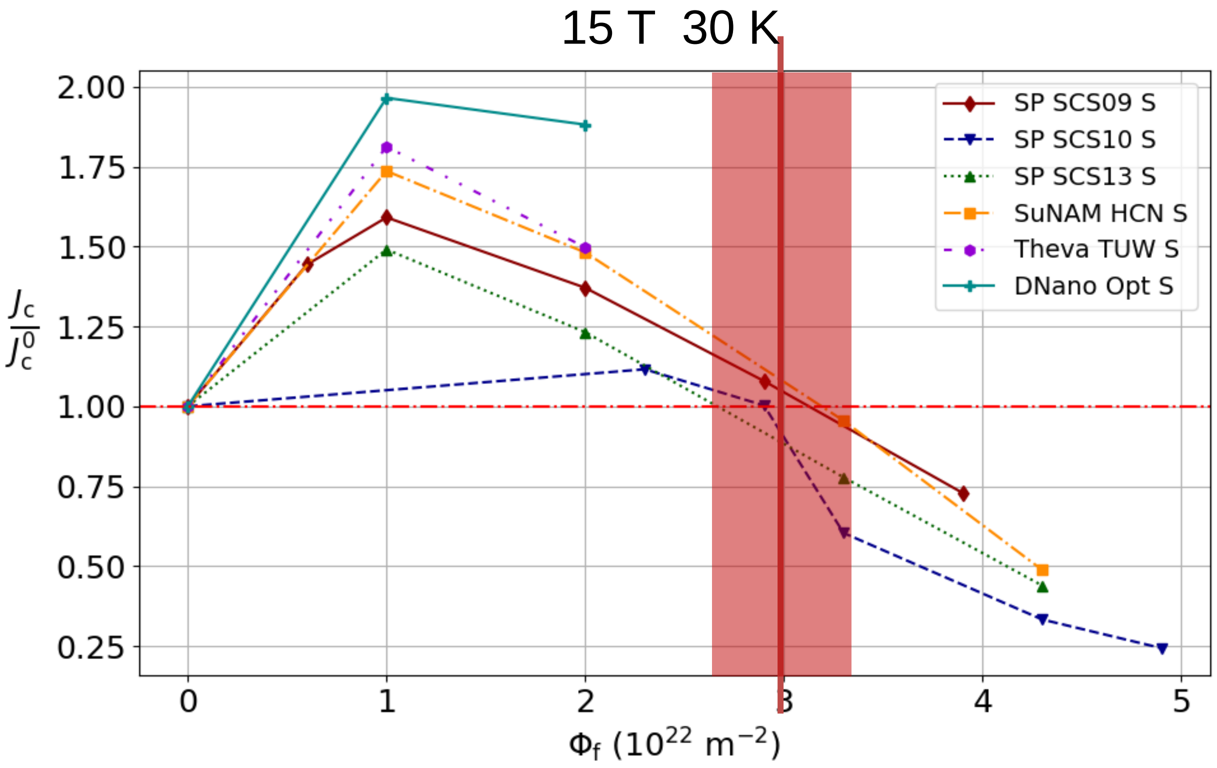
# Irradiation Damage

15 T  30 K



- Non-monotonic behavior of critical current as function of the neutron fluence
- Increase at low fluence
- Decrease at higher fluences

# Irradiation Damage

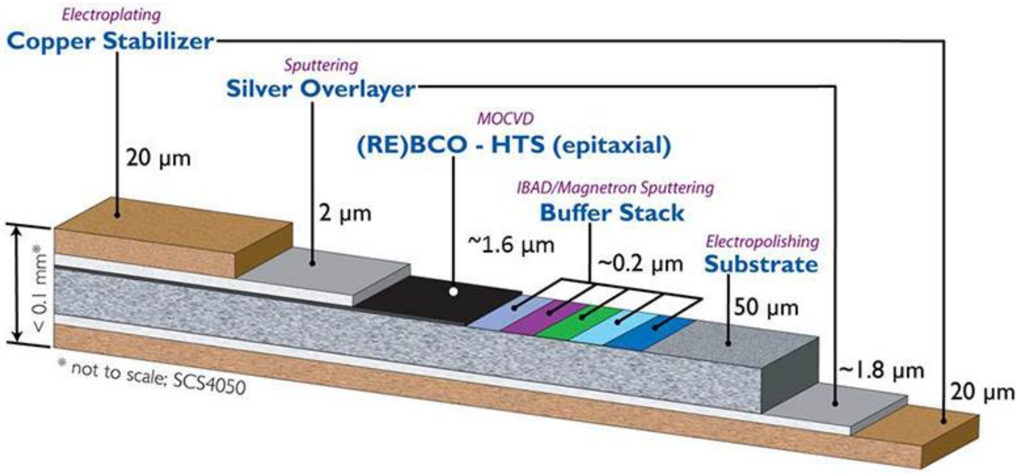


Compact devices

- Much higher neutron flux at the magnets
- Maximum lifte-time fluence of approx. 3-3.3 $\cdot 10^{22}$ m$^{-2}$

# Irradiation Methods

# Samples



[1] SuperPower®, superpower-inc.com

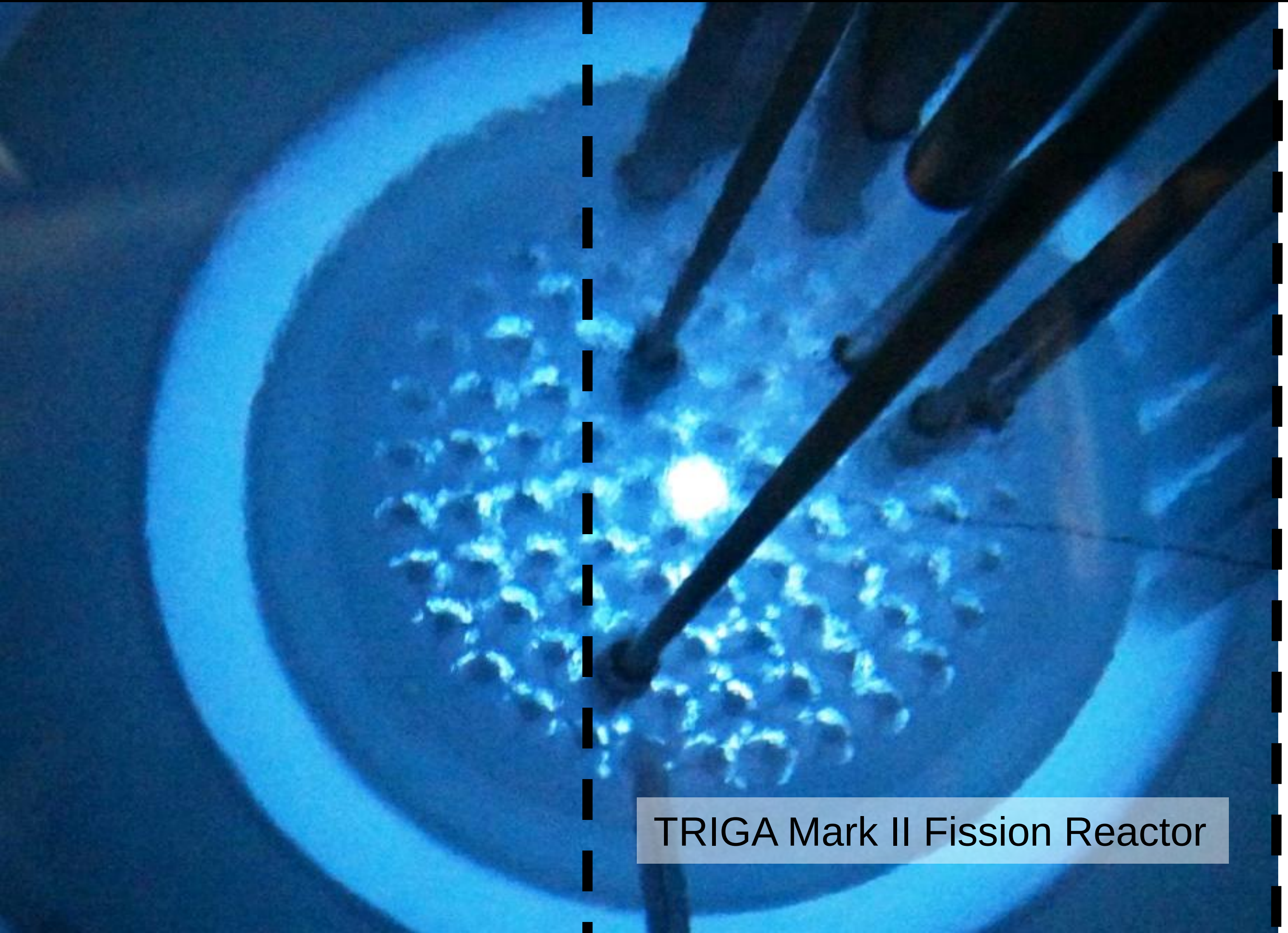
- chem. stabilized: 1 µm Ag
- el. stabilized: Cu
- substrate: Hastelloy
- HTS thickness: ~1 µm

**Thorough pre-characterization!**

| Supplier | Type | REBCO | APCs | method | nomenclature |
|---|---|---|---|---|---|
| SuperPower | SCS4050 2009 | GdBCO | None | MOCVD | SP SCS09 |
| SuperPower | SCS4050 2013 | (Y,Gd)BCO | $BaZrO_3$ | MOCVD | SP SCS13 |
| SuNAM | HCN04150 | GdBCO | None | RCE-DR | SuNAM HCN |

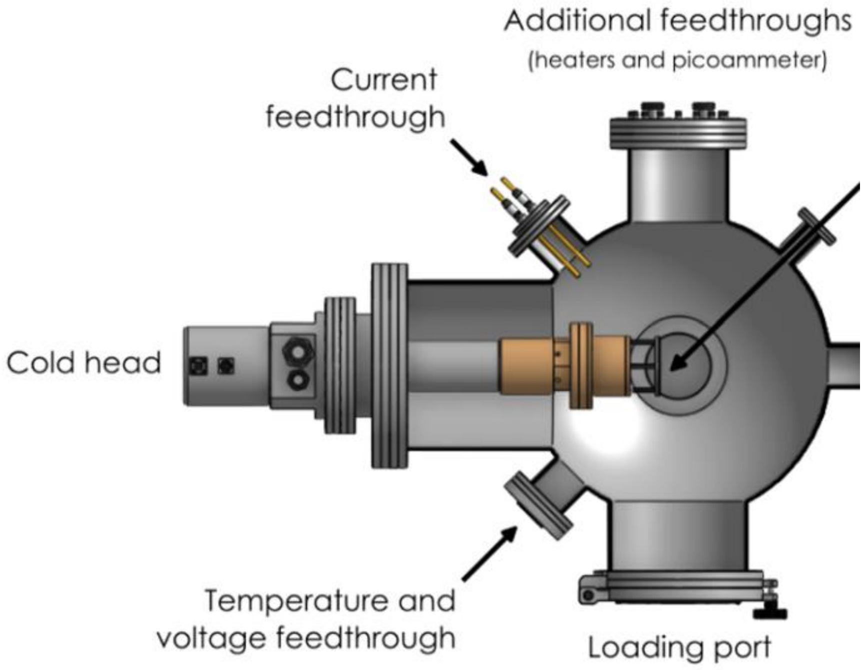
# Irradiation Environments

## Neutron irradiation at TU Wien



TRIGA Mark II Fission Reactor

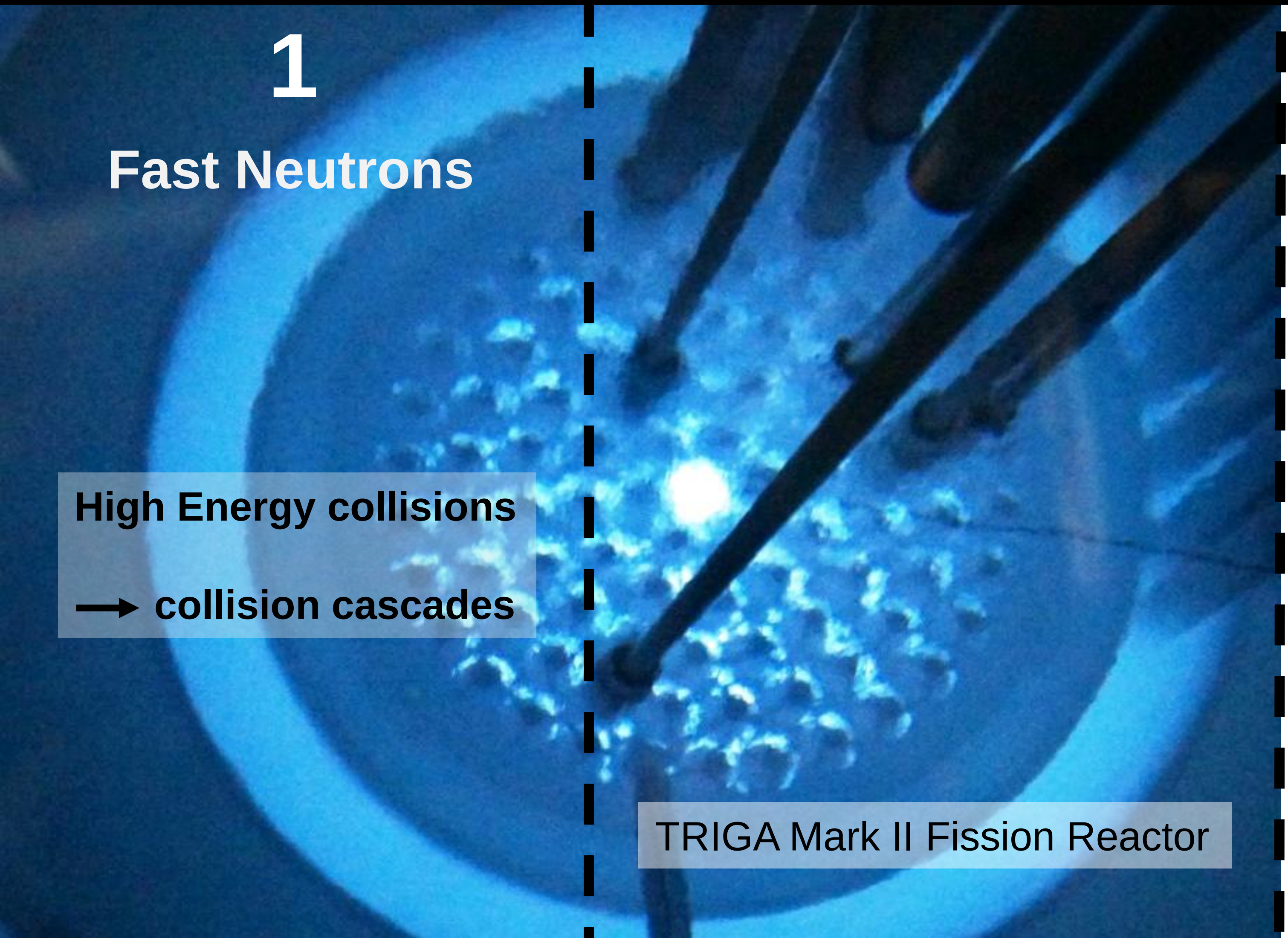
## Irradiation at MIT



General Ionix 1.7 MV Acc.

# Irradiation Environments

## Neutron irradiation at TU Wien



1

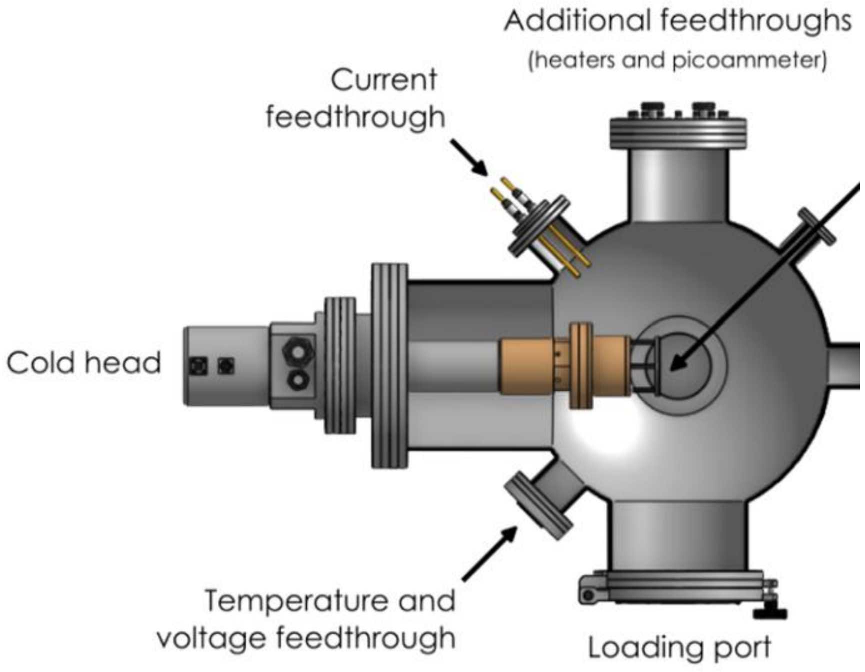
**Fast Neutrons**

**High Energy collisions**

**⟶ collision cascades**

TRIGA Mark II Fission Reactor

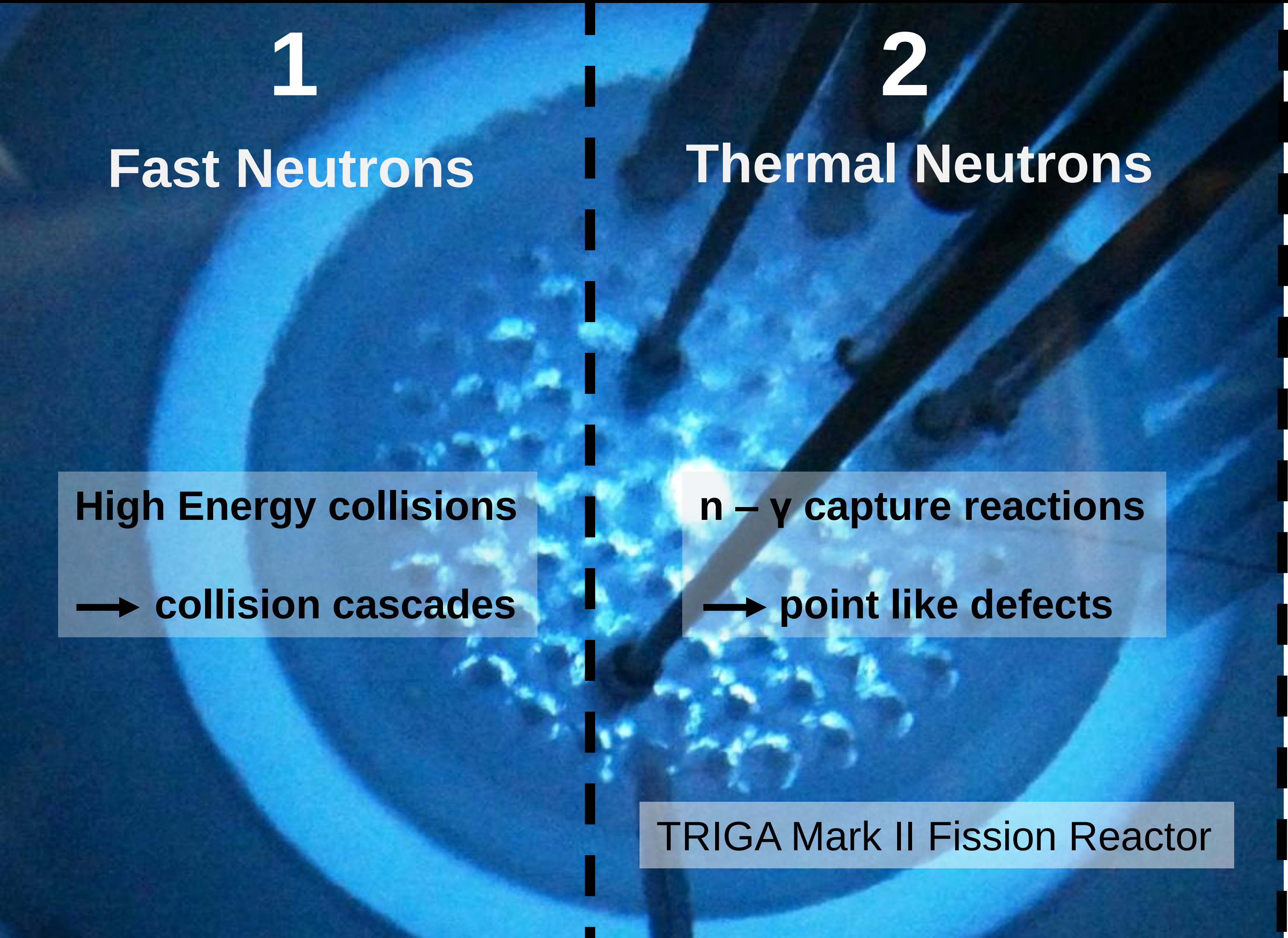
## Irradiation at MIT



General Ionix 1.7 MV Acc.

# Irradiation Environments

## Neutron irradiation at TU Wien



1

**Fast Neutrons**

**High Energy collisions**

**⟶ collision cascades**

2

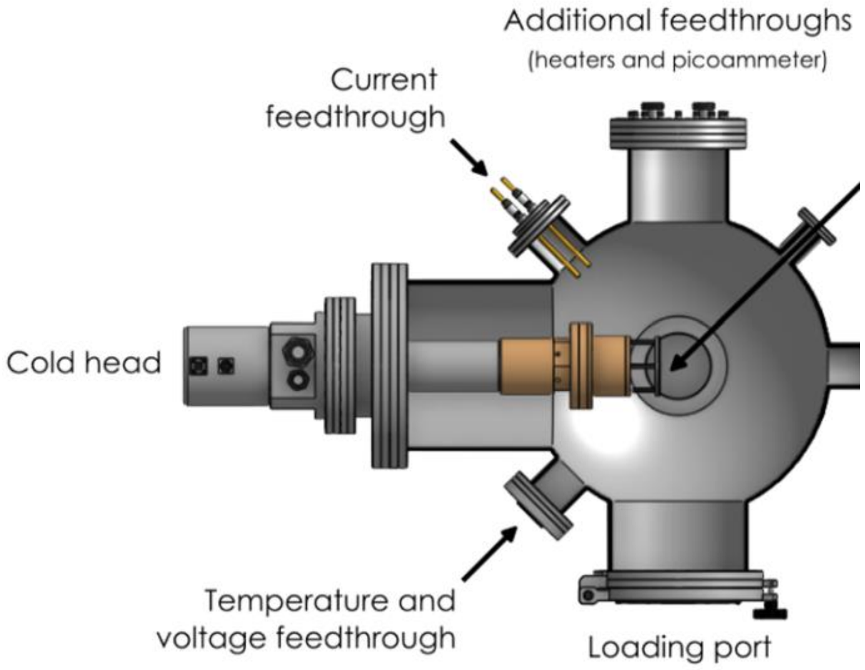
**Thermal Neutrons**

**n – γ capture reactions**

**⟶ point like defects**

TRIGA Mark II Fission Reactor

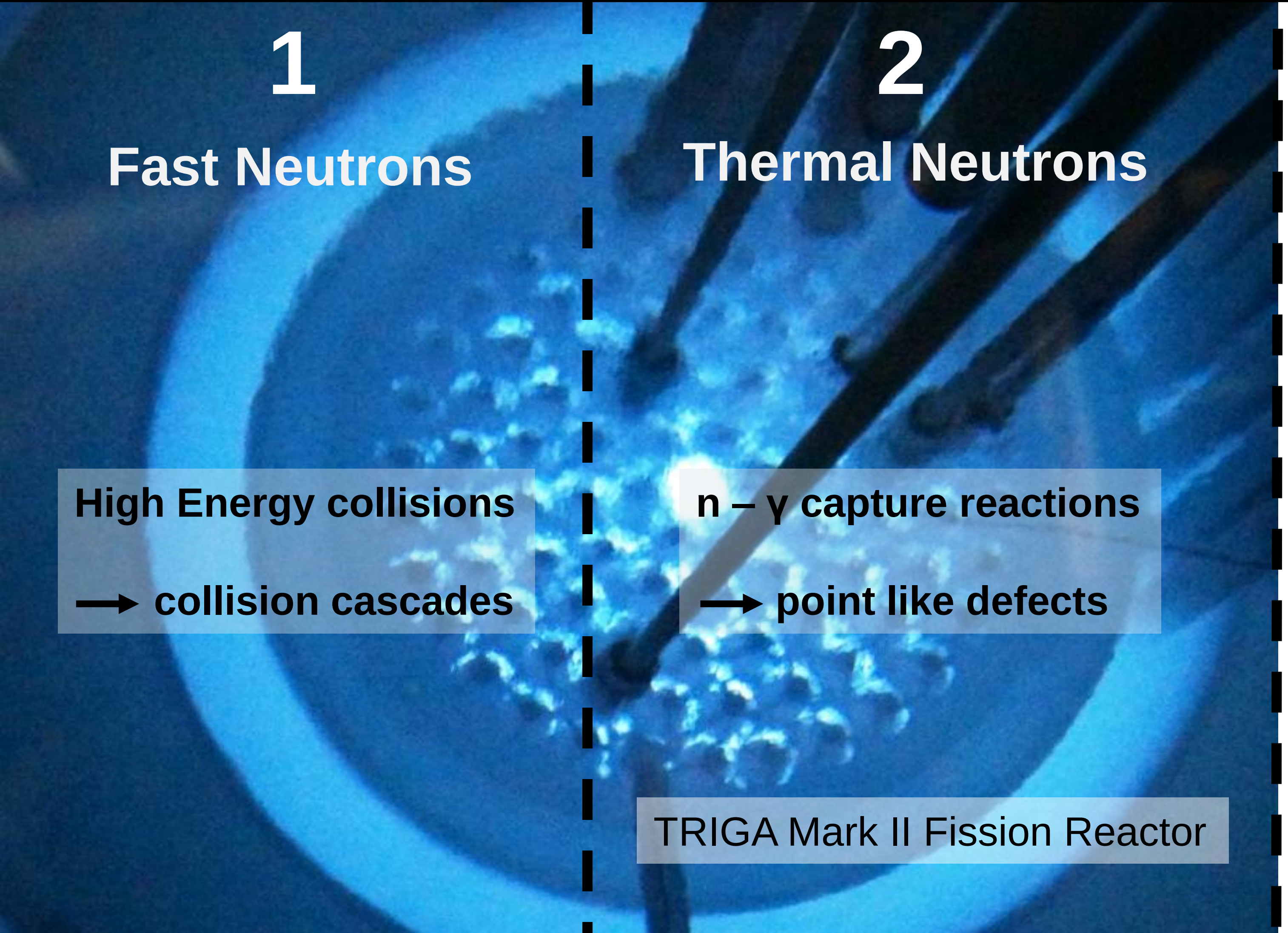
## Irradiation at MIT



General Ionix 1.7 MV Acc.

# Irradiation Environments

## Neutron irradiation at TU Wien



### 1

### Fast Neutrons

High Energy collisions

⟶ collision cascades

### 2

### Thermal Neutrons

n – γ capture reactions

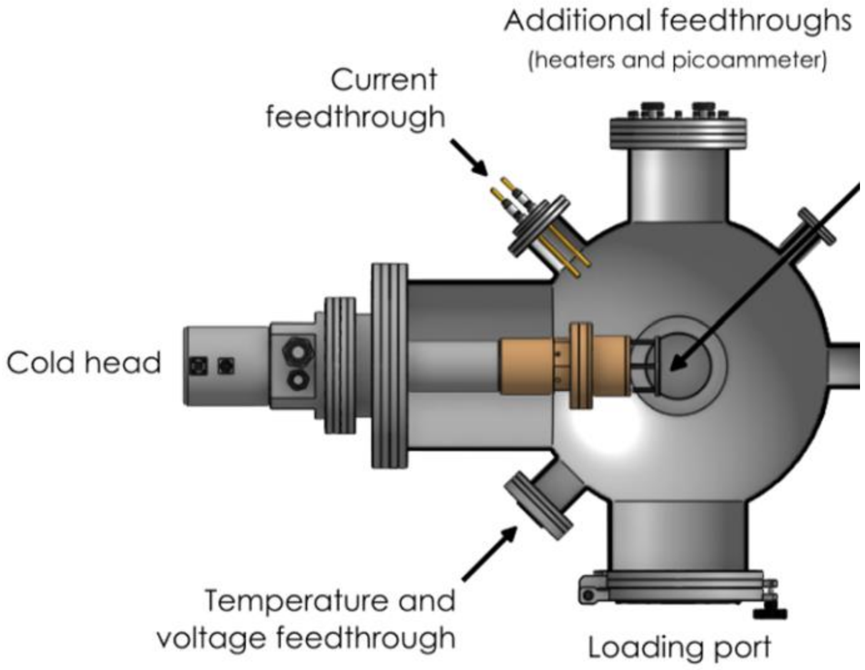
⟶ point like defects

TRIGA Mark II Fission Reactor

## Irradiation at MIT

### 3

### 1.2 MeV $p^+$ Control Experiment



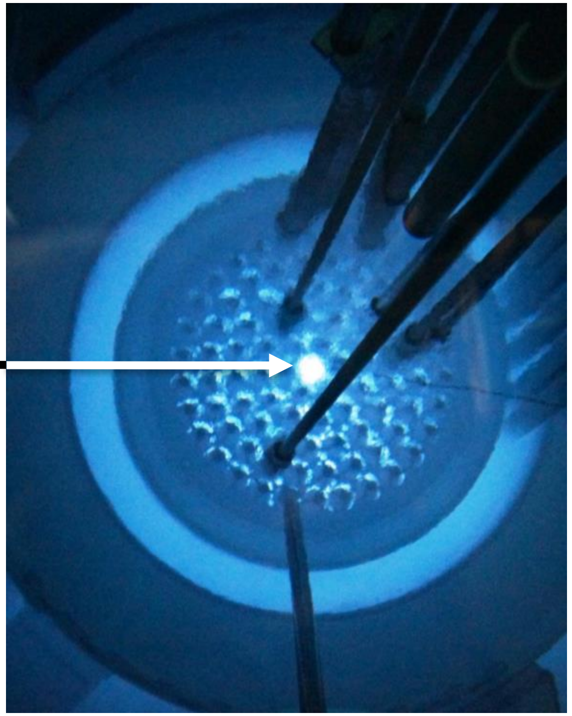
General Ionix 1.7 MV Acc.

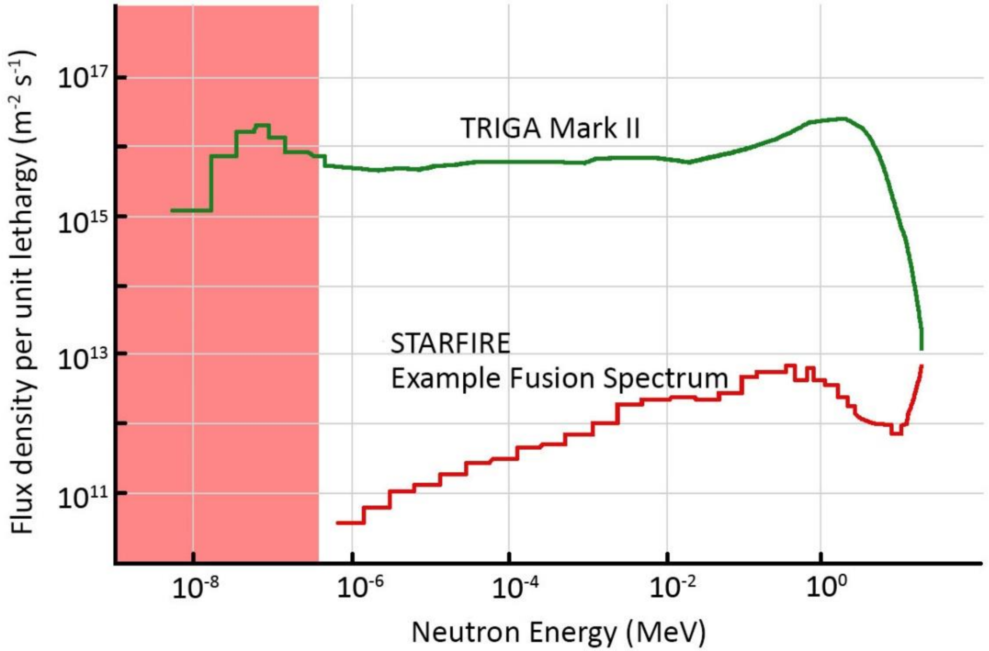
# Neutron Irradiation – Shielded

**TRIGA MARK II at TU Wien**

- Irradiation in the central irradiation facility
- Fast / thermal **neutron** flux 3.2 / 4 x $10^{16}$ $m^{-2}$ $s^{-1}$
- Irradiation with and **without** thermal (< 0.55 eV) neutrons
- Sample identifiers denoted with “S”

< 70 °C at sample



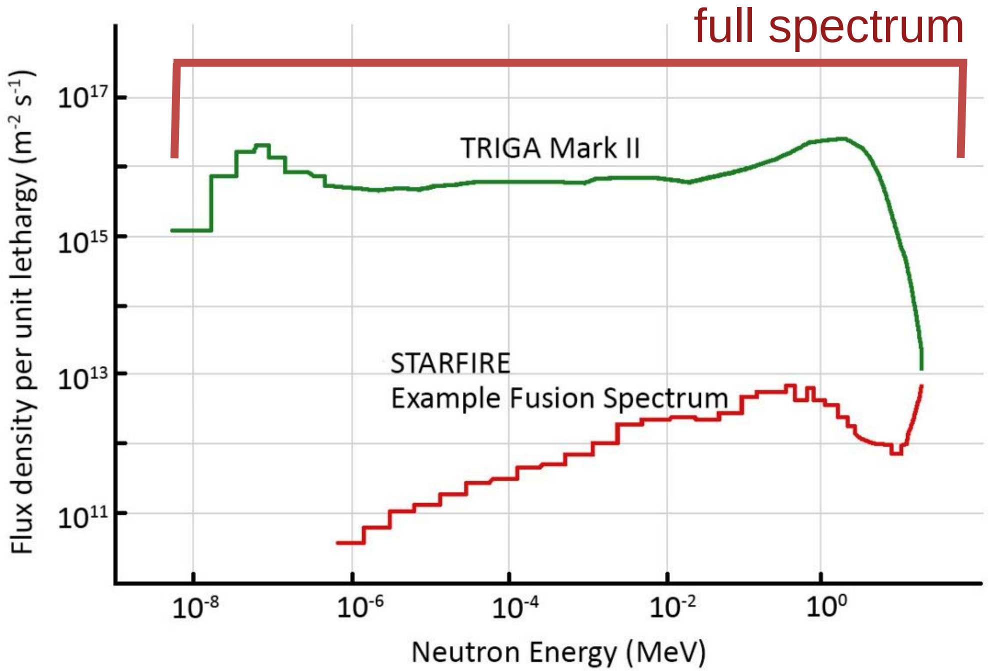


🟥 - can be shielded with Cd

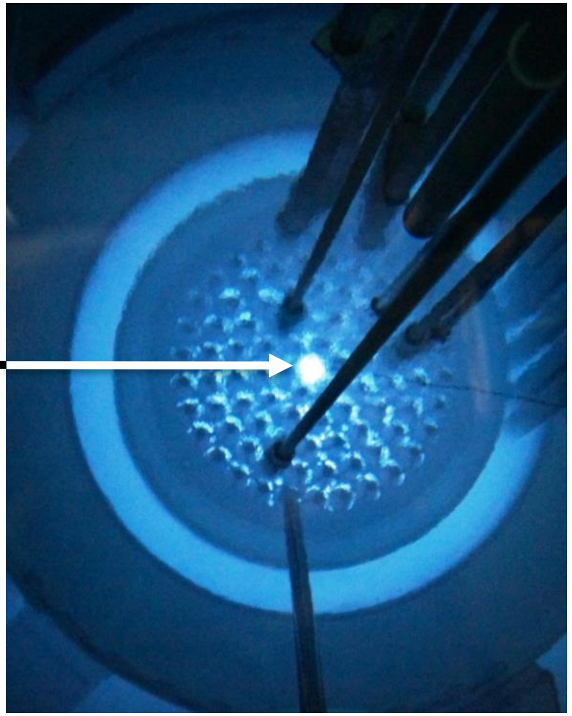
# Neutron Irradiation – Unshielded

**TRIGA MARK II at TU Wien**

- Irradiation in the central irradiation facility
- Fast / thermal **neutron** flux 3.2 / 4 x $10^{16}$ $m^{-2}$ $s^{-1}$
- Irradiation **with** and without thermal (< 0.55 eV) neutrons
- Sample identifiers denoted with "U"



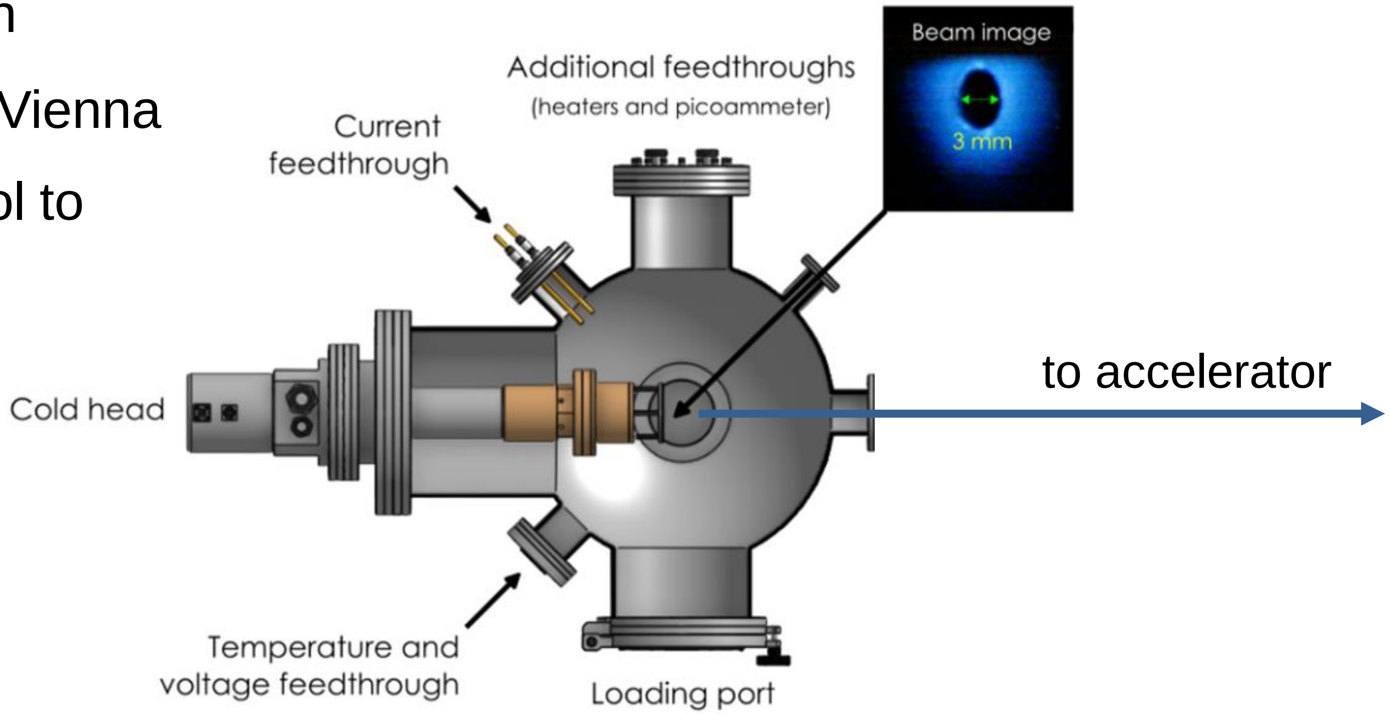
< 70 °C at sample



# $p^+$ Irradiation - Bridged

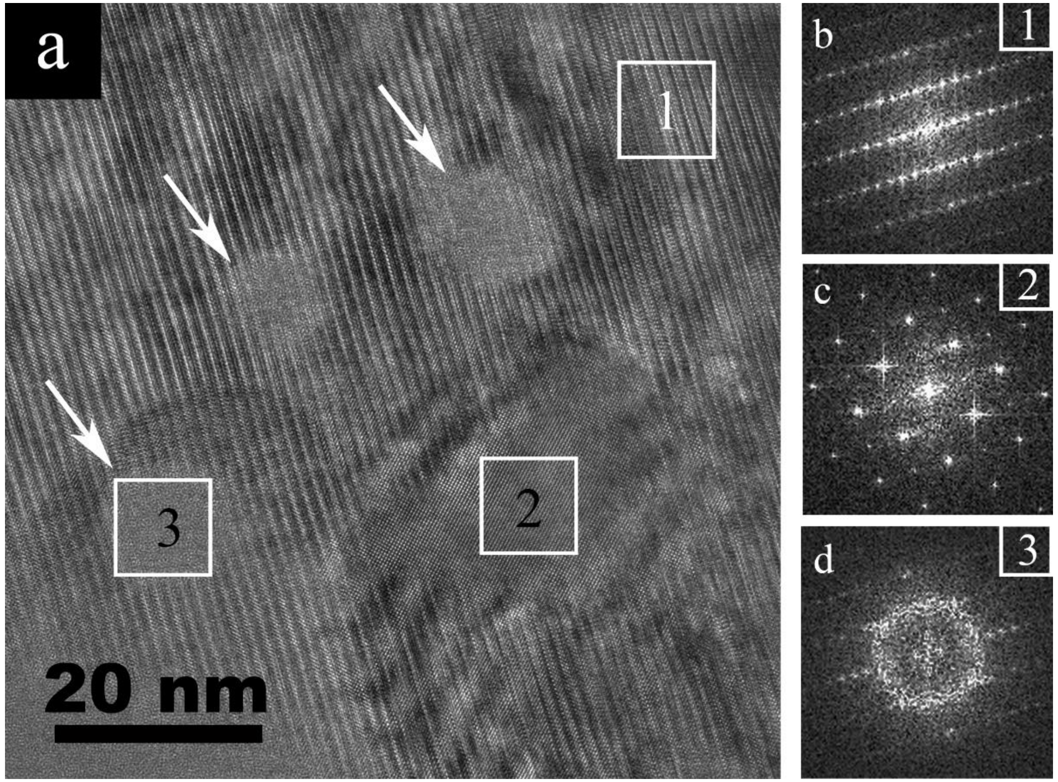
**General Ionix 1.7 MV tandem accelerator**

- Irradiation with 1.2 MeV protons
- Room temperature irradiation
- Bridged samples 0.2 mm width
- Samples pre-characterized in Vienna
- On-sample temperature control to monitor beam heating



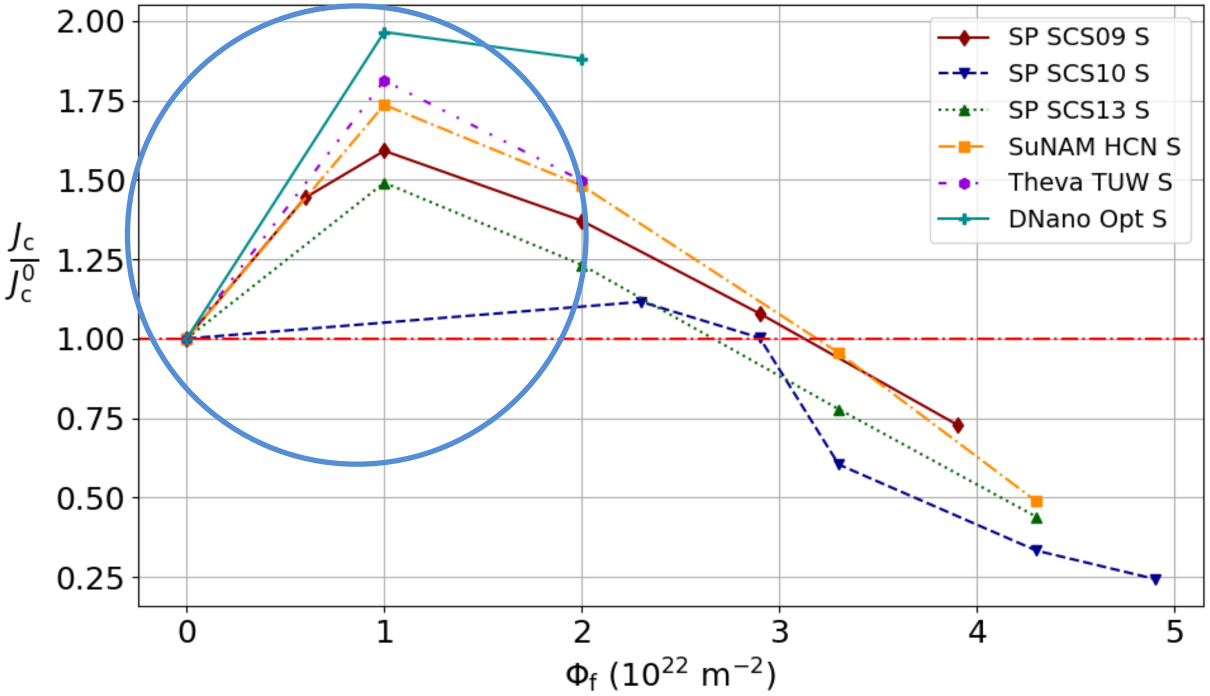
# Defect Formation

# Fast Neutron Irradiation



**left** – TEM picture of neutron induced defects
**right** – FFT of selected regions [1]

15 T  30 K

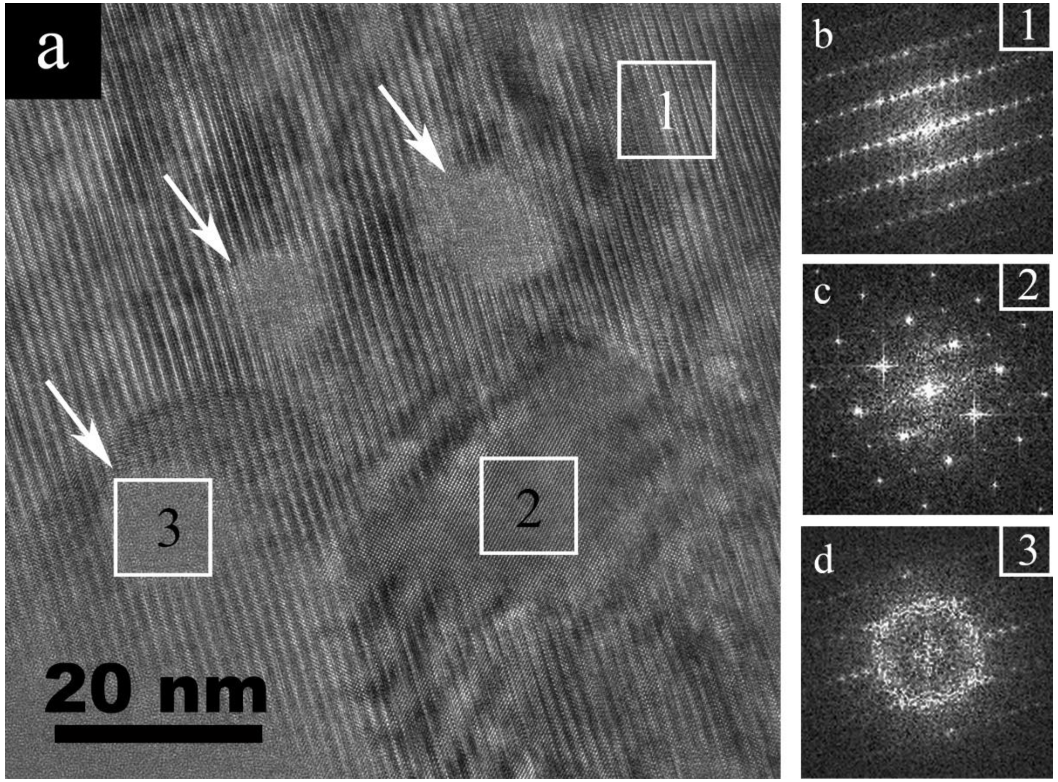


**Cascades Enhance Pinning**

---

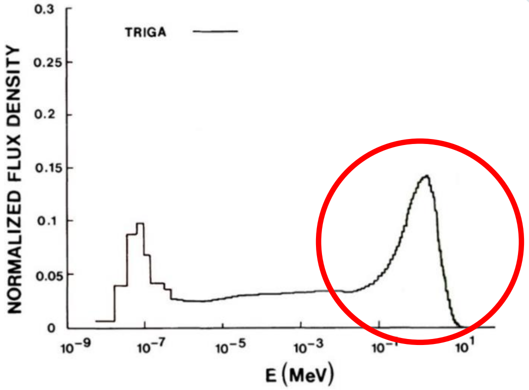
[1] with friendly permission by Yatir Linden, *Analysing neutron radiation damage in YBa2Cu3O7–x high-temperature superconductor tapes,* https://doi.org/10.1111/jmi.13078
Department of Materials, University of Oxford, Oxford, UK

# Fast Neutron Irradiation



**left** – TEM picture of neutron induced defects
**right** – FFT of selected regions [1]

1. Undisturbed GdBCO
2. Crystalline BZO rod
3. Amorphous cascade

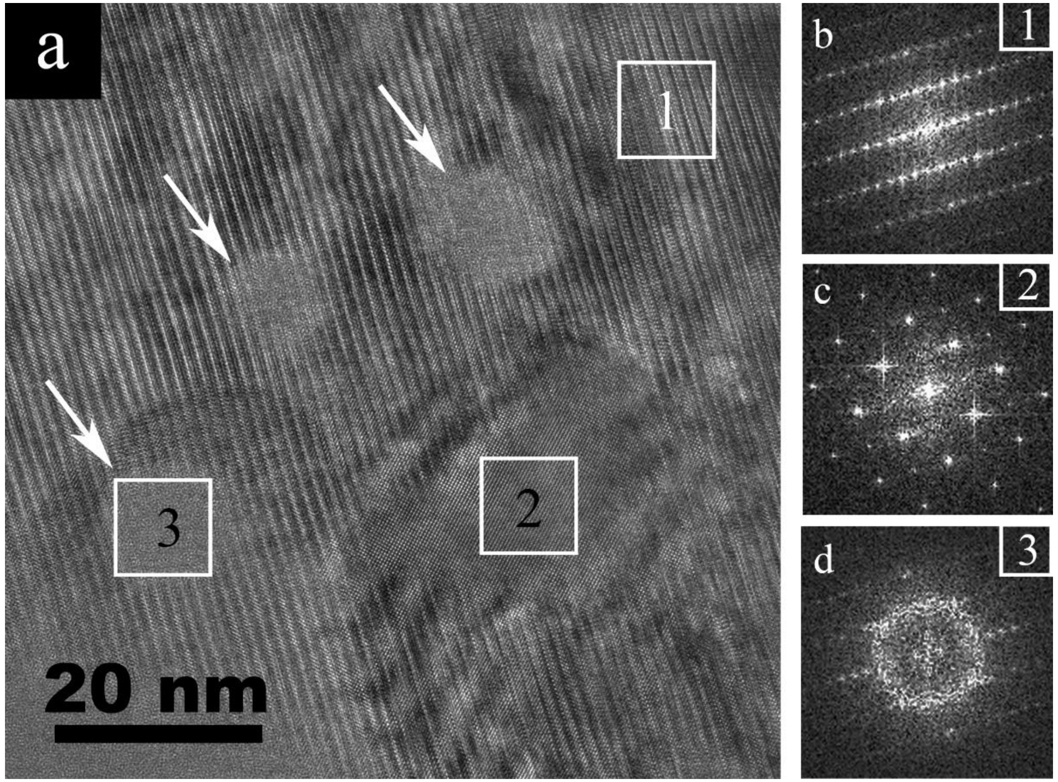


| | |
|---|---|
| Defect size | ≤ 10 nm |
| Mean | ~ 4 nm |
| $\xi^{0}_{ab}$ | ~ 1.4 nm |
| $\xi^{77}_{ab}$ | ~ 3 nm |

**Cascades Enhance Pinning**

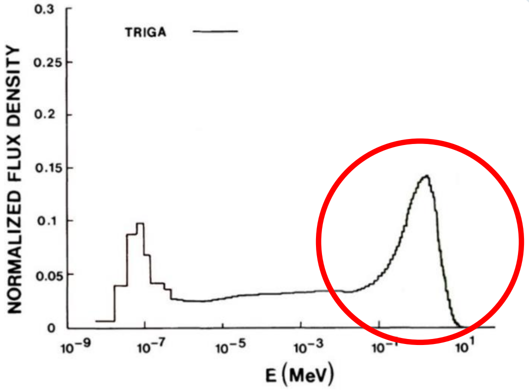
[1] with friendly permission by Yatir Linden, *Analysing neutron radiation damage in YBa2Cu3O7–x high-temperature superconductor tapes,* https://doi.org/10.1111/jmi.13078
Department of Materials, University of Oxford, Oxford, UK

# Fast Neutron Irradiation



**left** – TEM picture of neutron induced defects
**right** – FFT of selected regions [1]

1. Undisturbed GdBCO
2. Crystalline BZO rod
3. Amorphous cascade



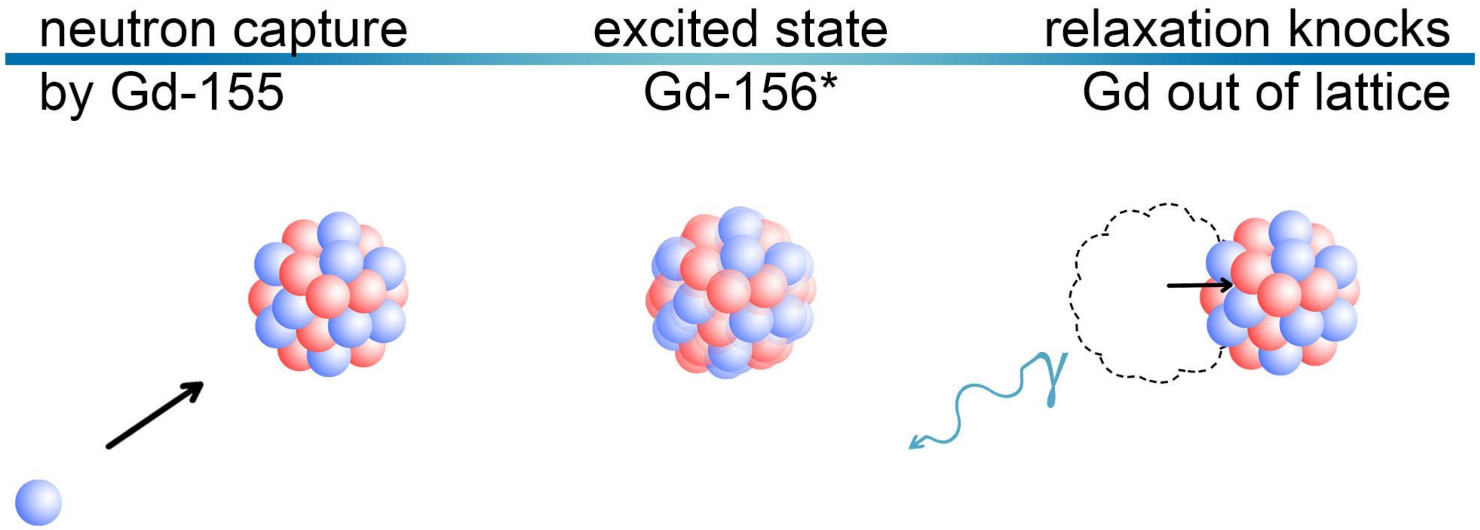
- $3.3 \times 10^{19} – 5 \times 10^{22}$ cascades per $10^{22}$
- ~ 0.01 % reduction of superconducting cross-section

**What drives the degradation?**

➡ **Must be small (invisible) defects**

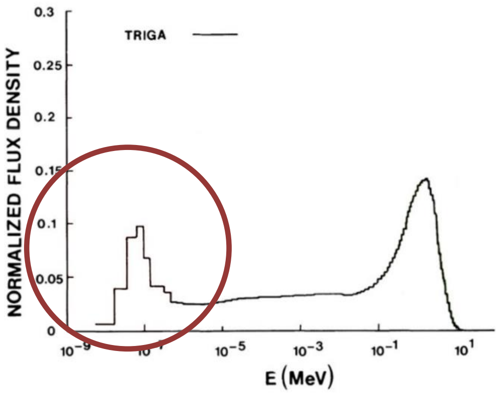
[1] with friendly permission by Yatir Linden, *Analysing neutron radiation damage in YBa2Cu3O7–x high-temperature superconductor tapes,* https://doi.org/10.1111/jmi.13078
Department of Materials, University of Oxford, Oxford, UK

# Thermal Neutron Irradiation


neutron capture by Gd-155

excited state Gd-156*

relaxation knocks Gd out of lattice

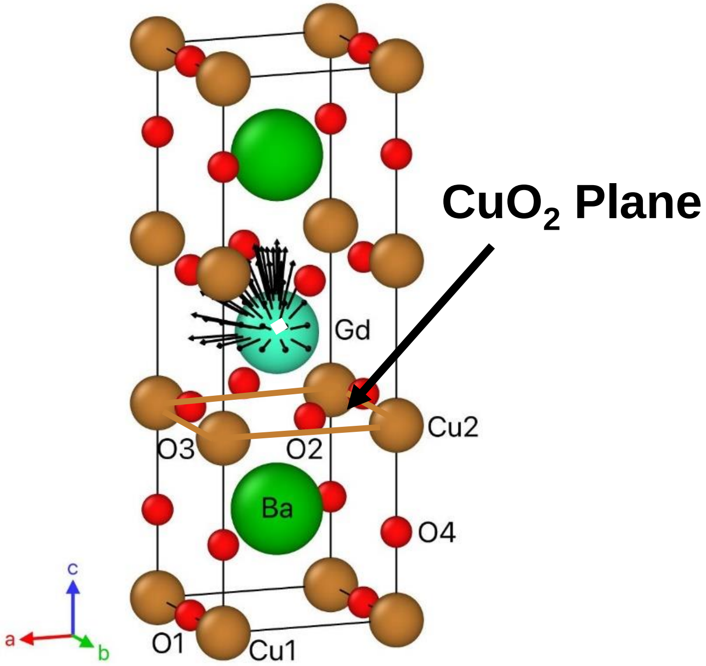


K.E. Sickafus et al., Phys. Rev. B **46** (1992) 11862

Thermal neutrons excite Gd → Recoil of 29 – 32 eV gamma emission displaces the nucleus

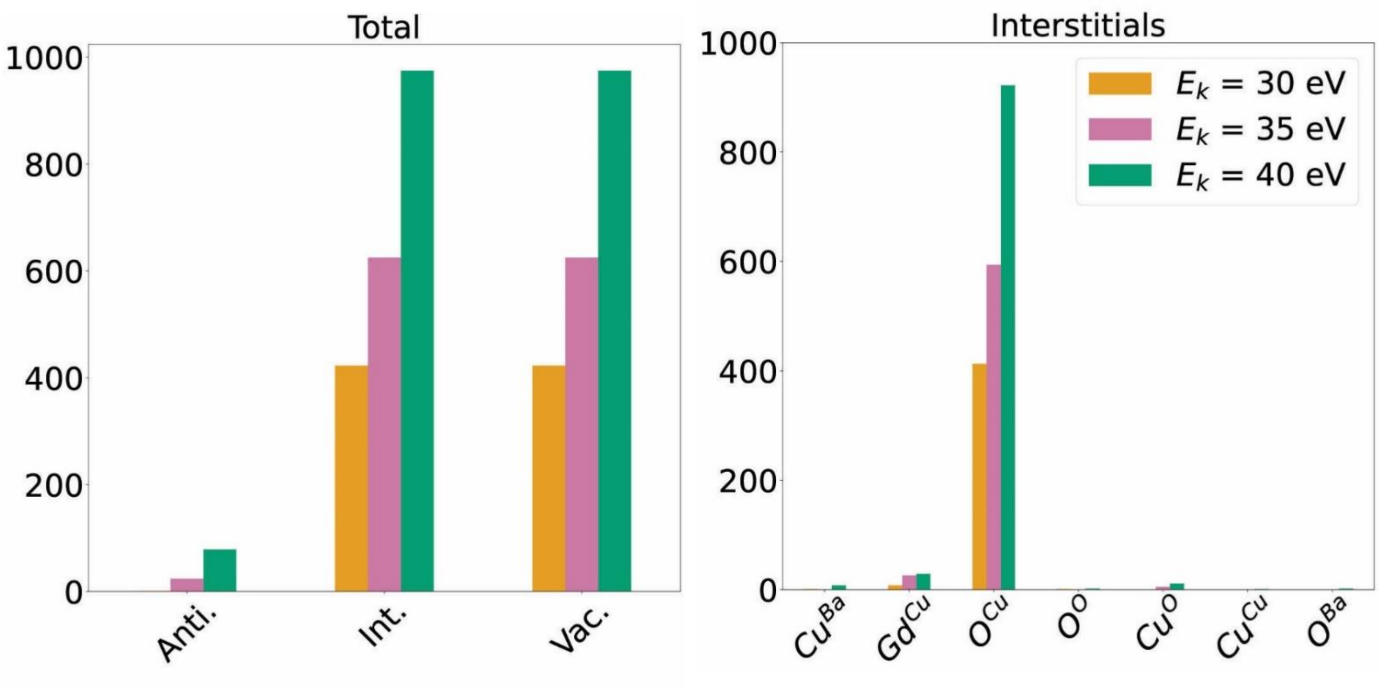
- Very high defect densities achievable
- Add to fast neutron induced defects

# Thermal Neutron Irradiation



- Position enables introduction of many defects close to the planes
- Defects are small in comparison to coll. cascades
- Defects may be modelled with MDS
- 3 energies close to experimental value simulated (30, 35, 40 eV)

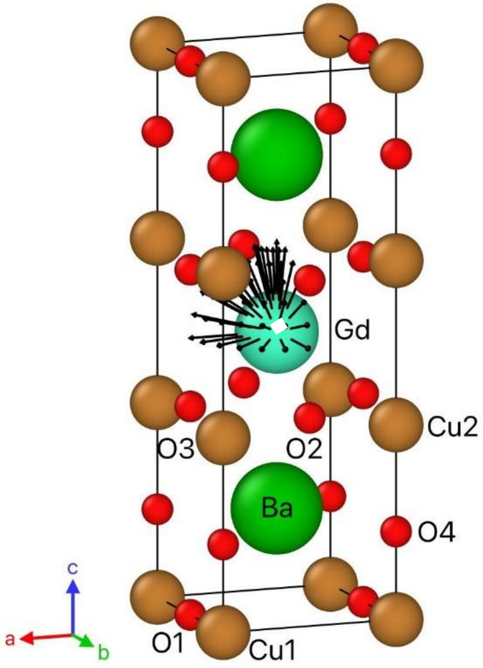
# Thermal Neutron Irradiation - MDS

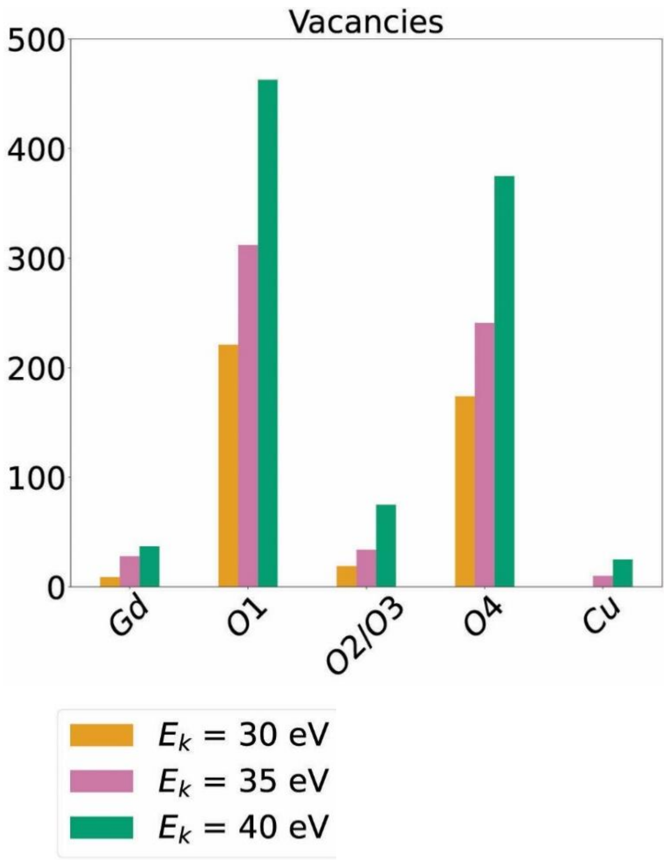


430 simulation runs per energy

- Most defects are oxygen vacancies
- Gd returns / stays in lattice position
- Different defects originating from Gd PKA (primary knock on atom)
- Defect distribution changes with energy
- 1-2 defects per incident particle

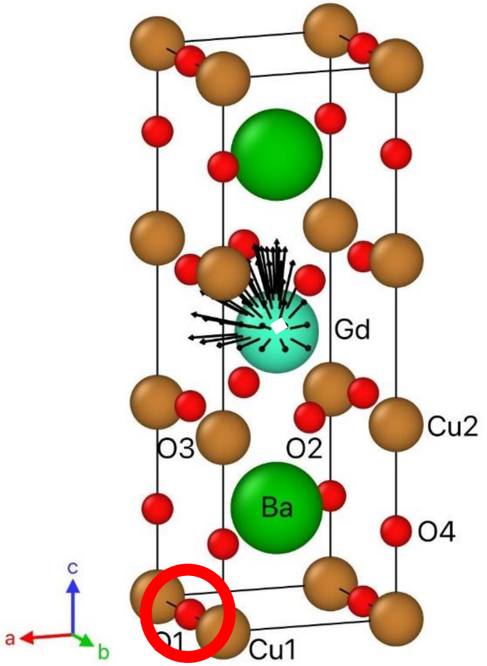
# Thermal Neutron Irradiation - MDS

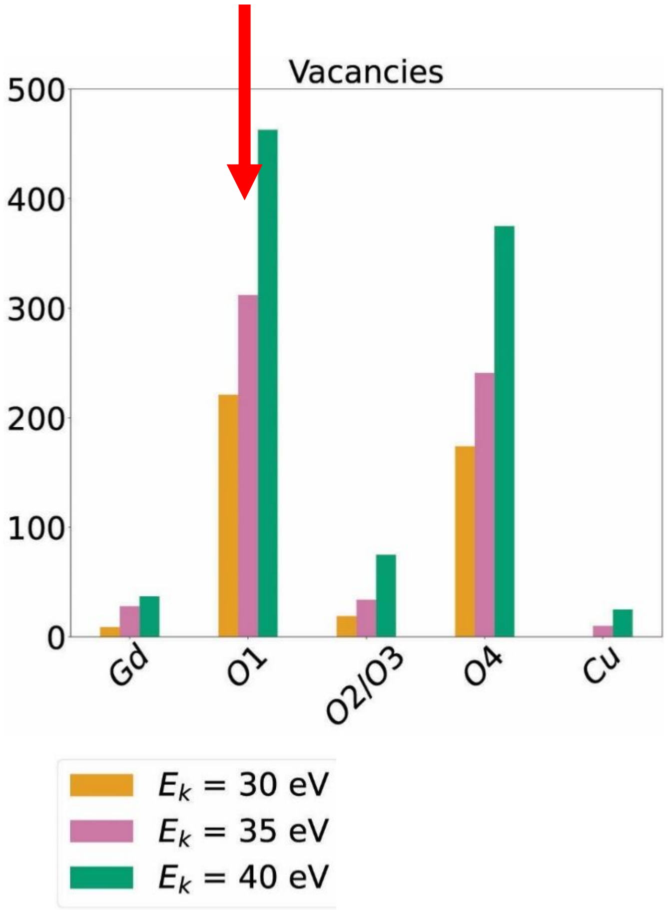




- Most defects are oxygen vacancies
- Gd returns / stays in lattice position
- Different defects originating from Gd PKA (primary knock on atom)
- Defect distribution changes with energy
- 1-2 defects per incident particle

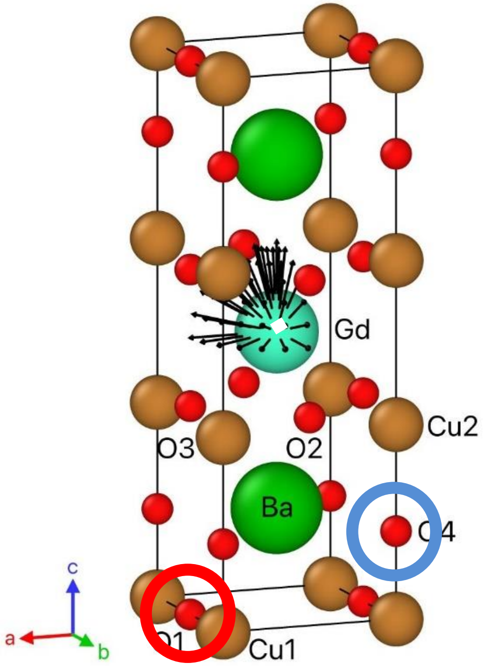
# Thermal Neutron Irradiation - MDS

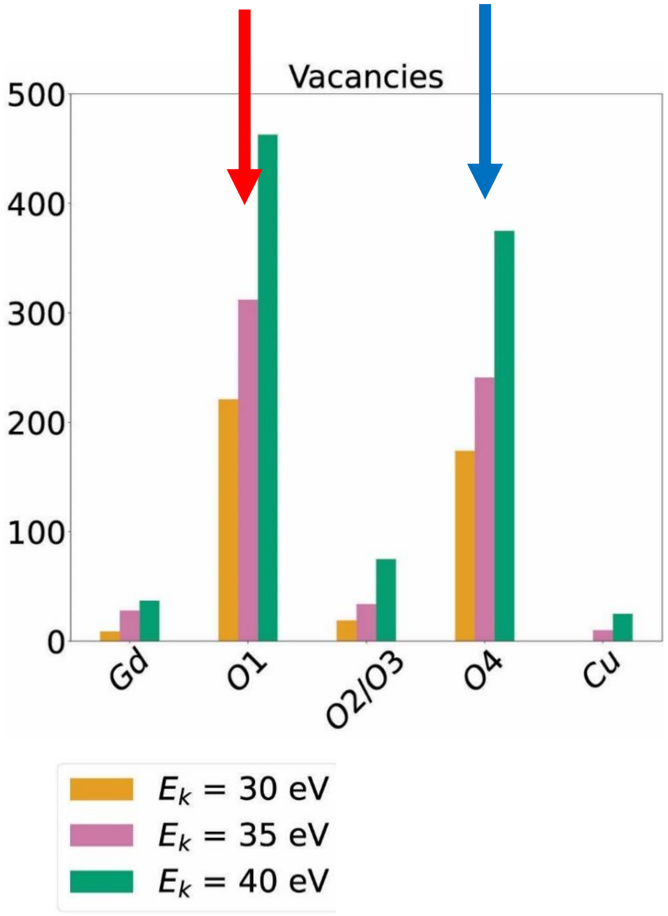




- Most defects are oxygen vacancies
- Gd returns / stays in lattice position
- Different defects originating from Gd PKA (primary knock on atom)
- Defect distribution changes with energy
- 1-2 defects per incident particle

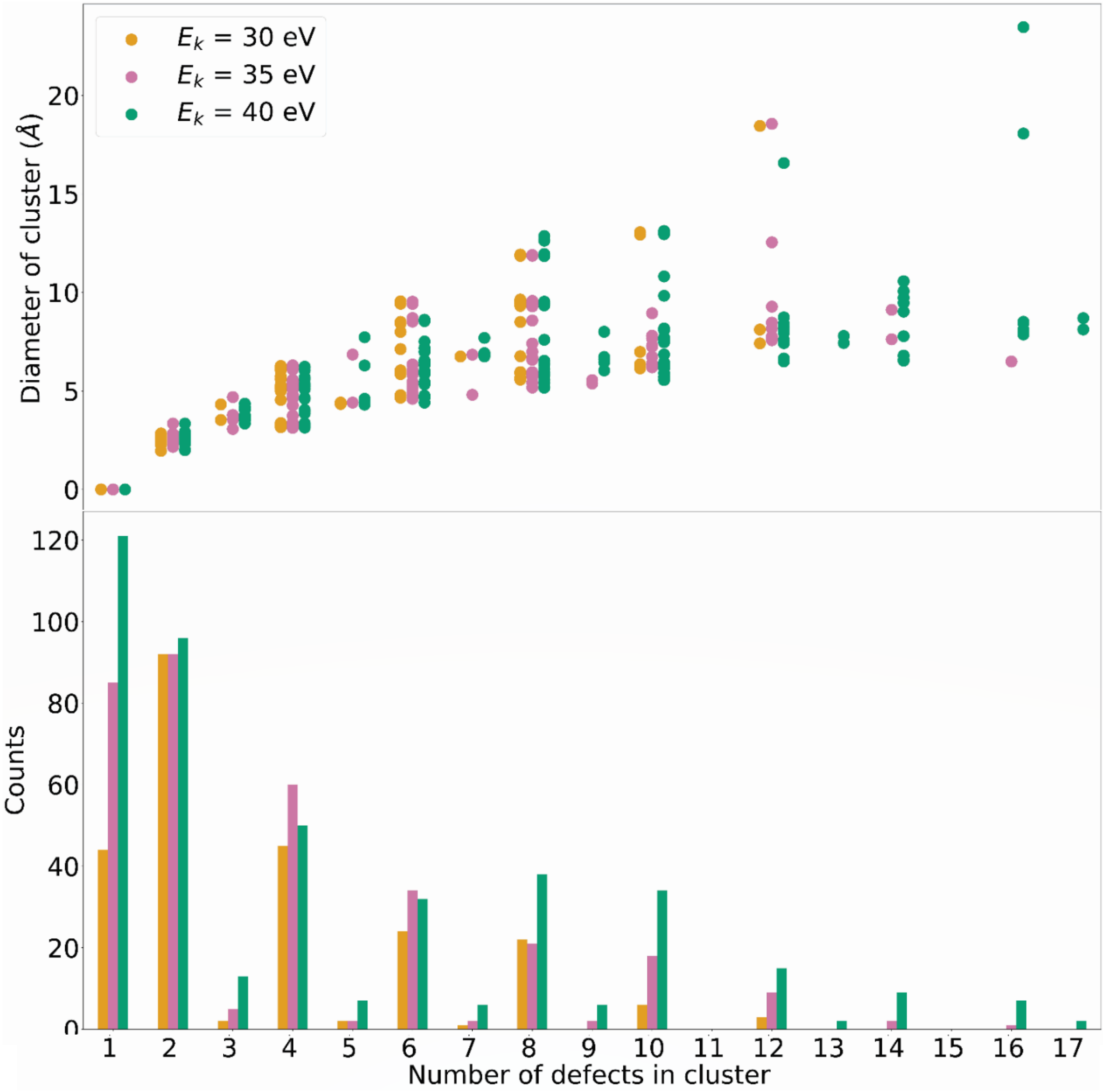
# Thermal Neutron Irradiation - MDS





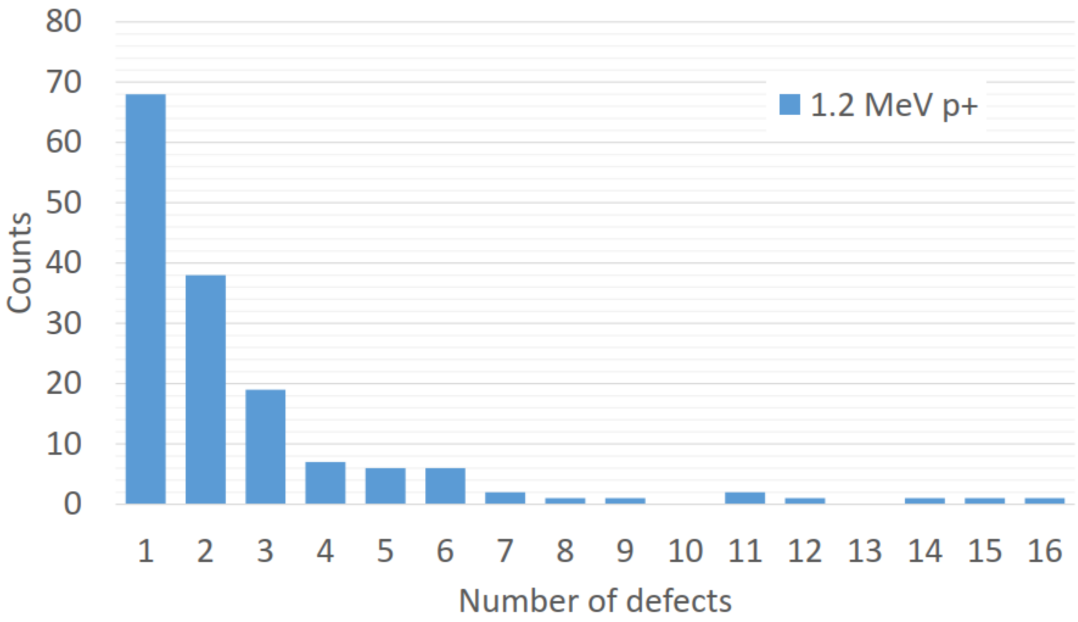
- Most defects are oxygen vacancies
- Gd returns / stays in lattice position
- Different defects originating from Gd PKA (primary knock on atom)
- Defect distribution changes with energy
- 1-2 defects per incident particle

# Thermal Neutron Irradiation - MDS



- Mainly point-like defects
- Small defects form clusters
- Up to 1 nm in size
- Slightly improves pinning behavior

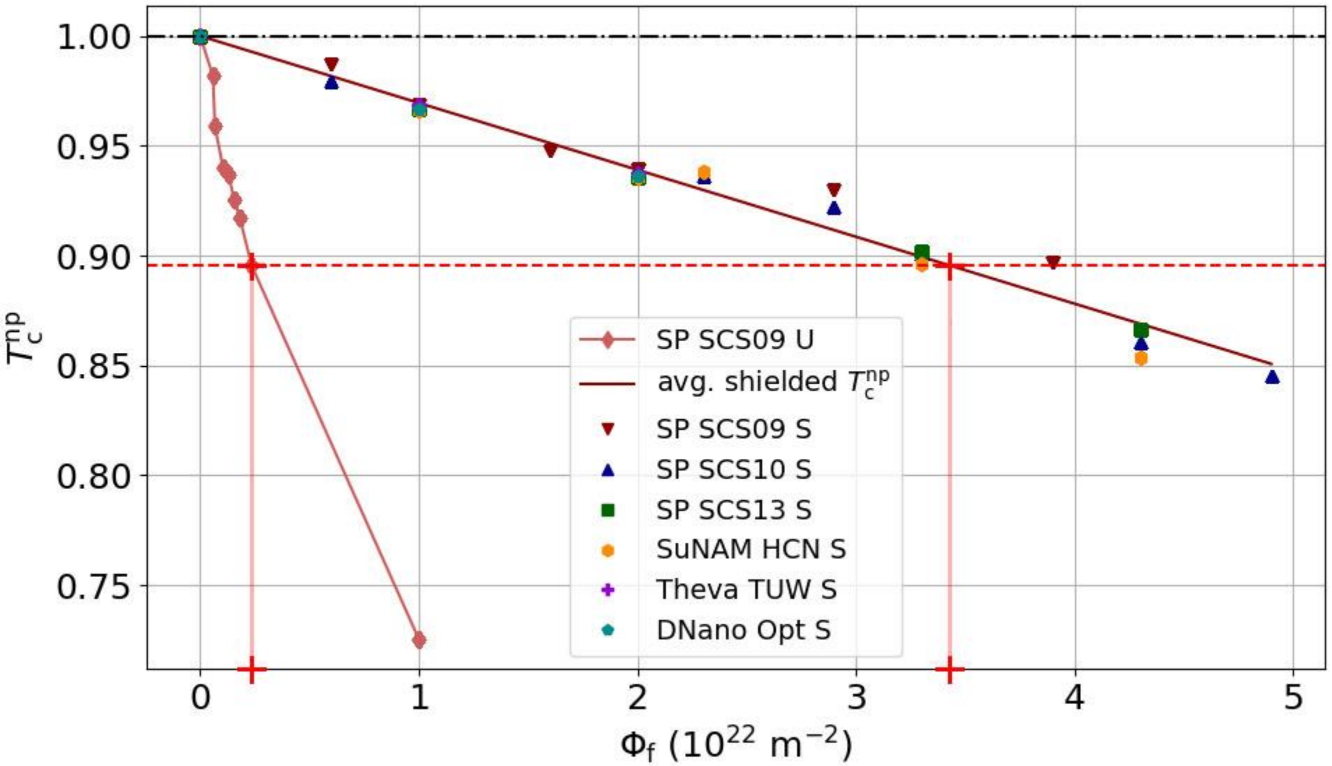
# 1.2 MeV $p^+$ Irradiation



- SRIM/TRIM
- Most defects are oxygen displacements (low binding energy)
- Little is known about actual defects and recombination
- Large defects are possible but rare
- Most defects are point-like or small clusters like with thermal neutrons

# Results

# Influence of thermal neutrons - $T_c$



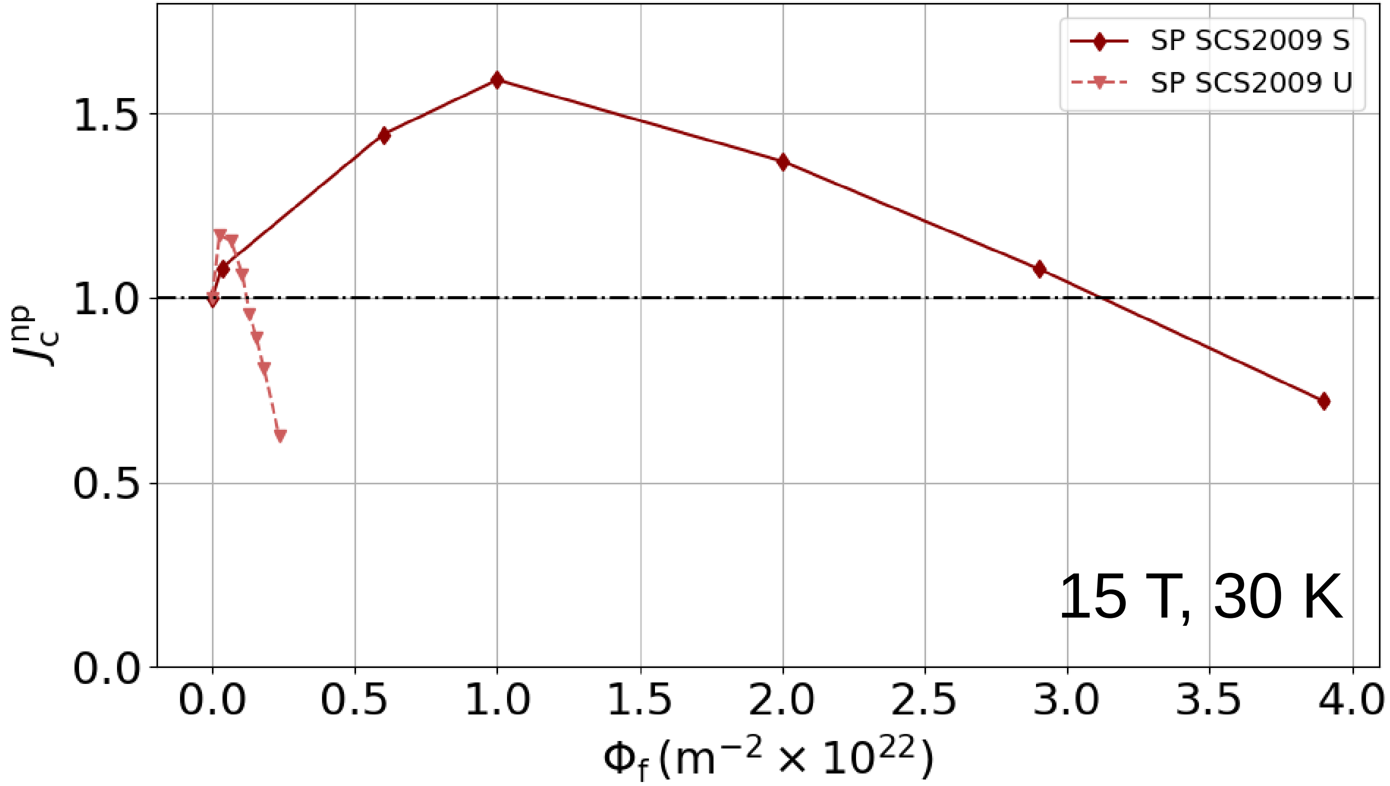
$$\frac{T_c}{T_c^0} = T_c^{\mathbf{np}}$$

**np**… normalized to pristine value

$T_c$ degrades ~14 x faster due to Gd-point defects

(MARS calculations: dpa only higher by about 50%)

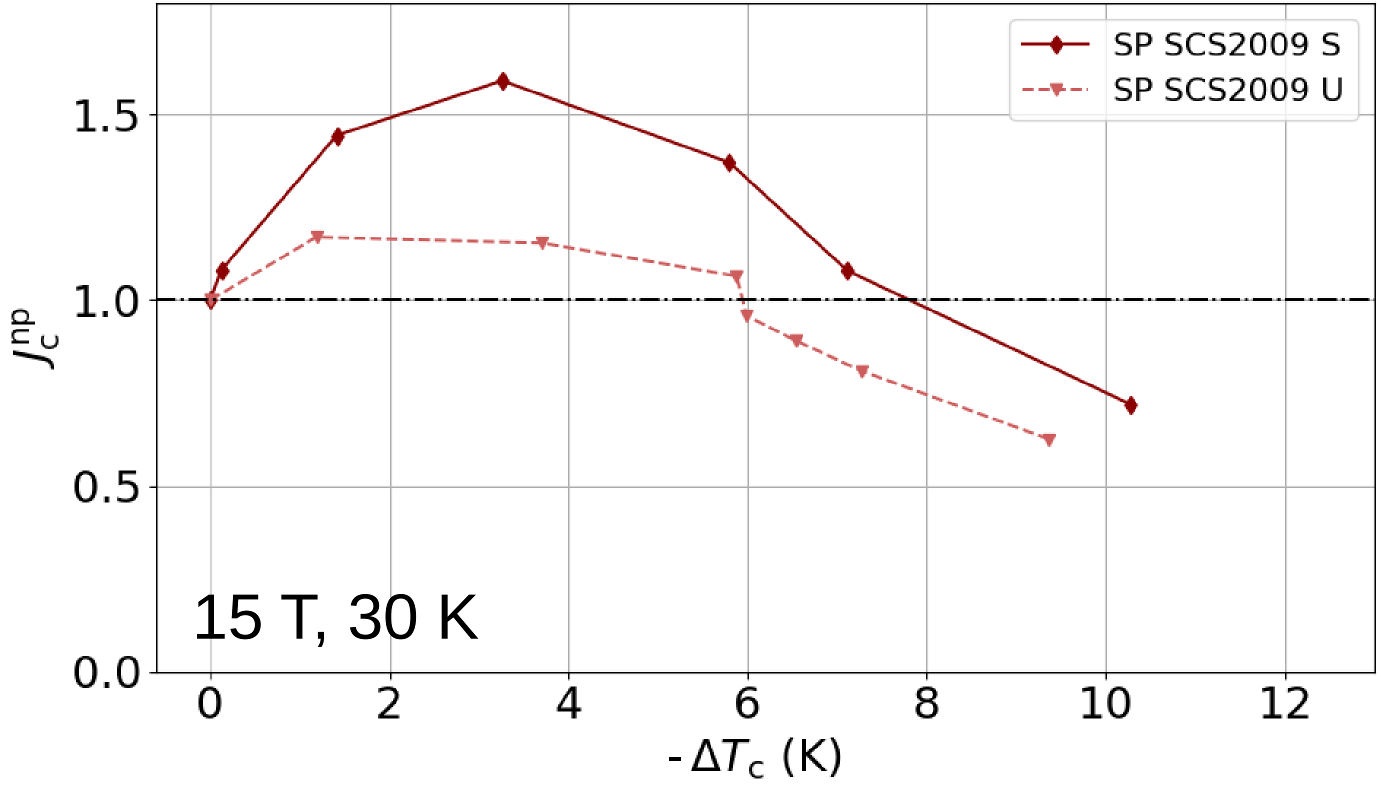
# Influence of thermal neutrons - $J_c$



- Maximum occurs at much lower neutron fluences
- $J_c$ at maximum is smaller
- Degradation is much faster

Fluence is not a good measure for the disorder!

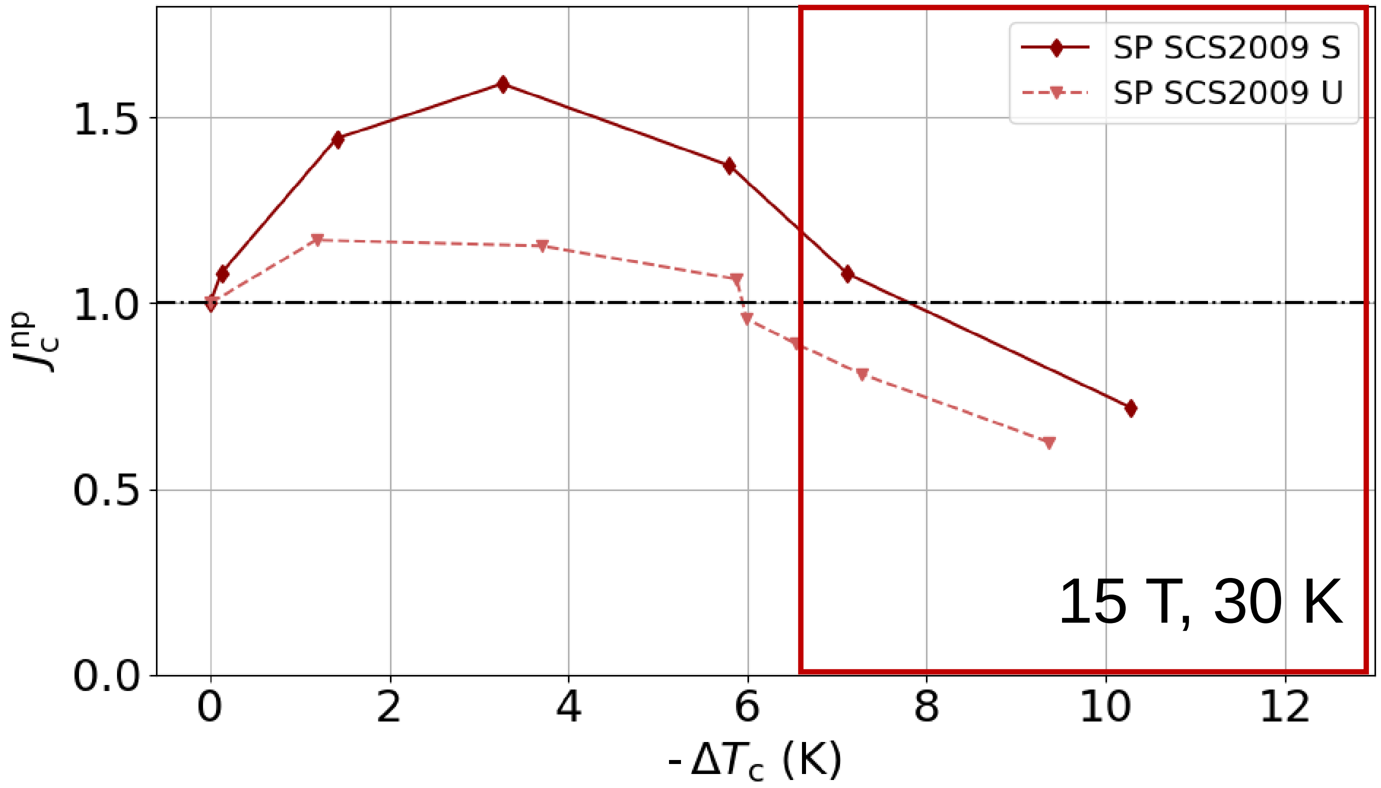
# Influence of thermal neutrons - $J_c$



- $J_c$ maximum shifted to lower $T_c$
- Degradation with similar slope
  - Accumulation of similar defects?

- $T_c$ is efficient disorder parameter (decrease of superfluid density)
- $D = T_c - T_c^p$

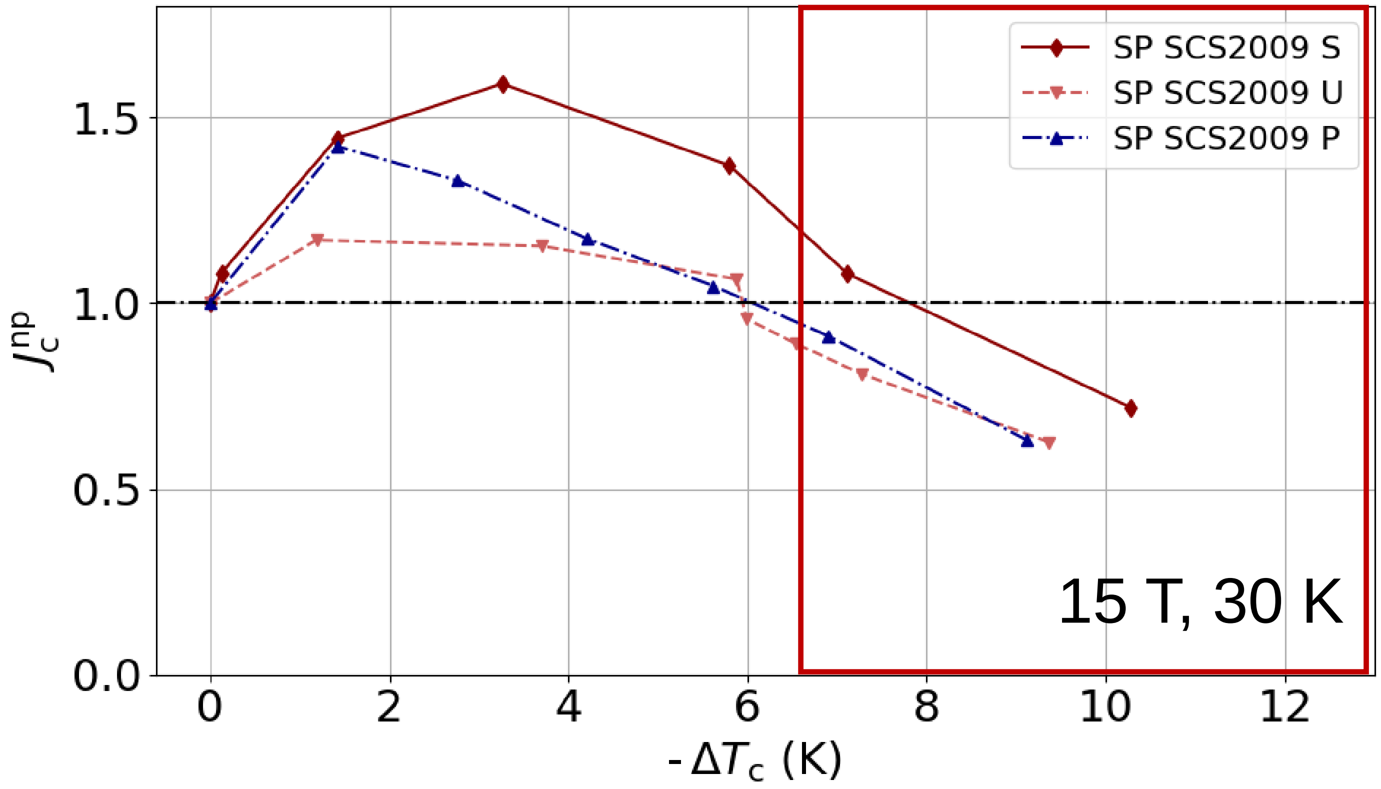
# Influence of thermal neutrons - $J_c$



- Different defect densities
- Parallel degrading branch
- Specific defects origin of degradation?
- Accumulation in all irradiation techniques?

**Focus on degrading branch**

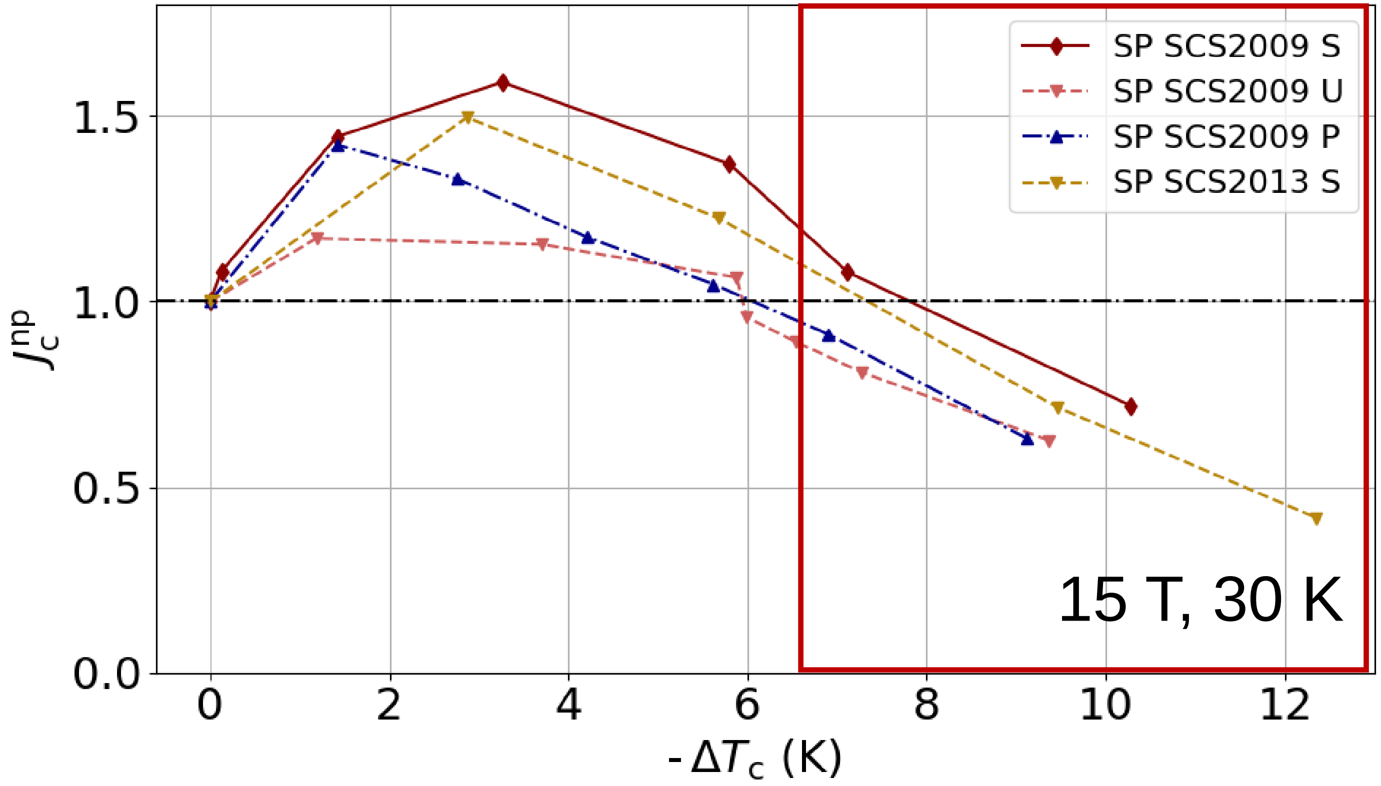
# Influence of thermal neutrons - $J_c$



- Different defect densities
- Parallel degrading branch
- Specific defects origin of degradation?
- Accumulation in all irradiation techniques?

**— · Sample irradiated with 1.2 MeV proton at room temperature**

# Influence of thermal neutrons - $J_c$

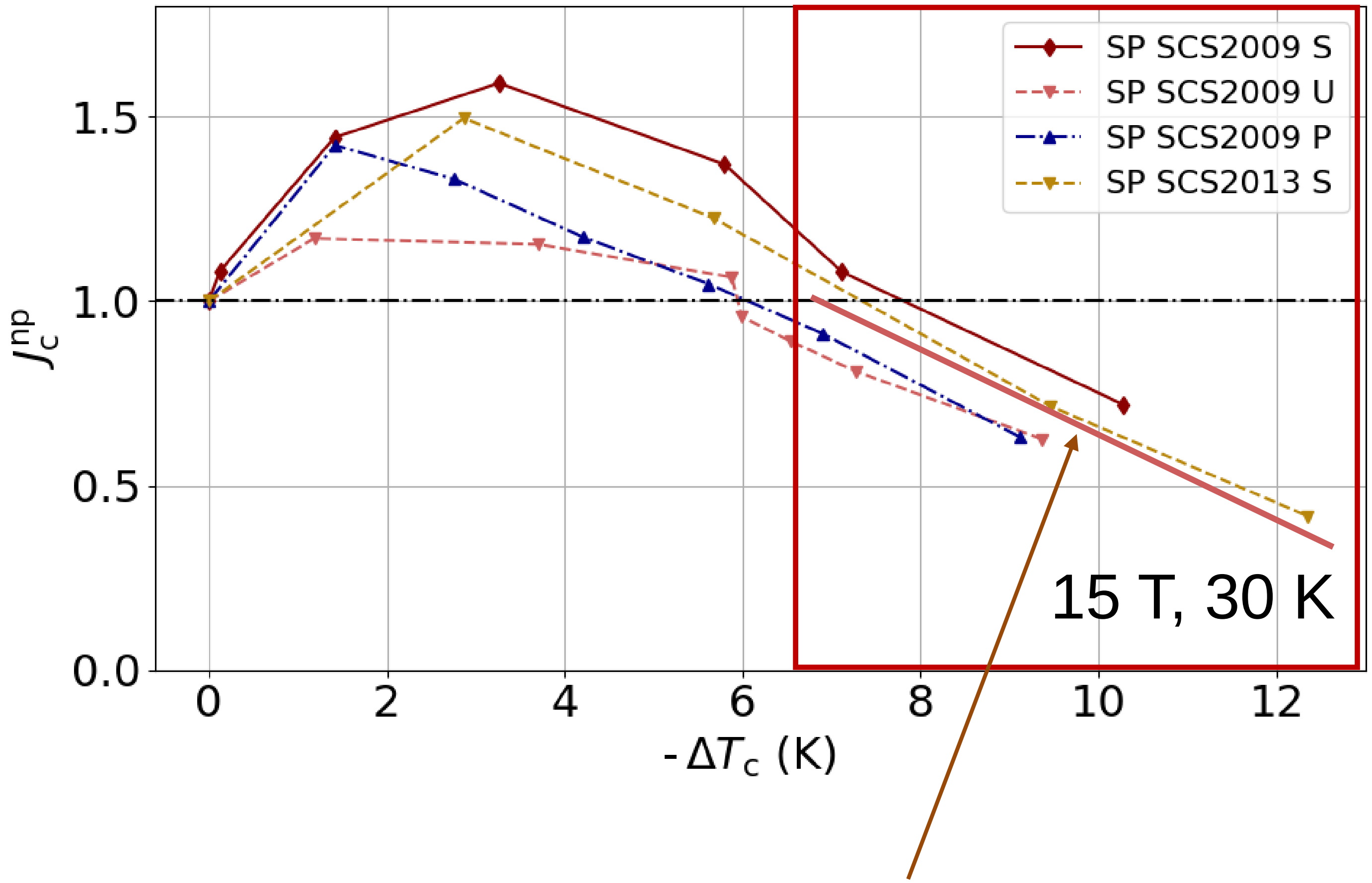


- Different defect densities
- Parallel degrading branch
- Specific defects origin of degradation?
- Accumulation in all irradiation techniques?

**Sample irradiated with 1.2 MeV proton at room temperature**

**Shielded sample with APCs**

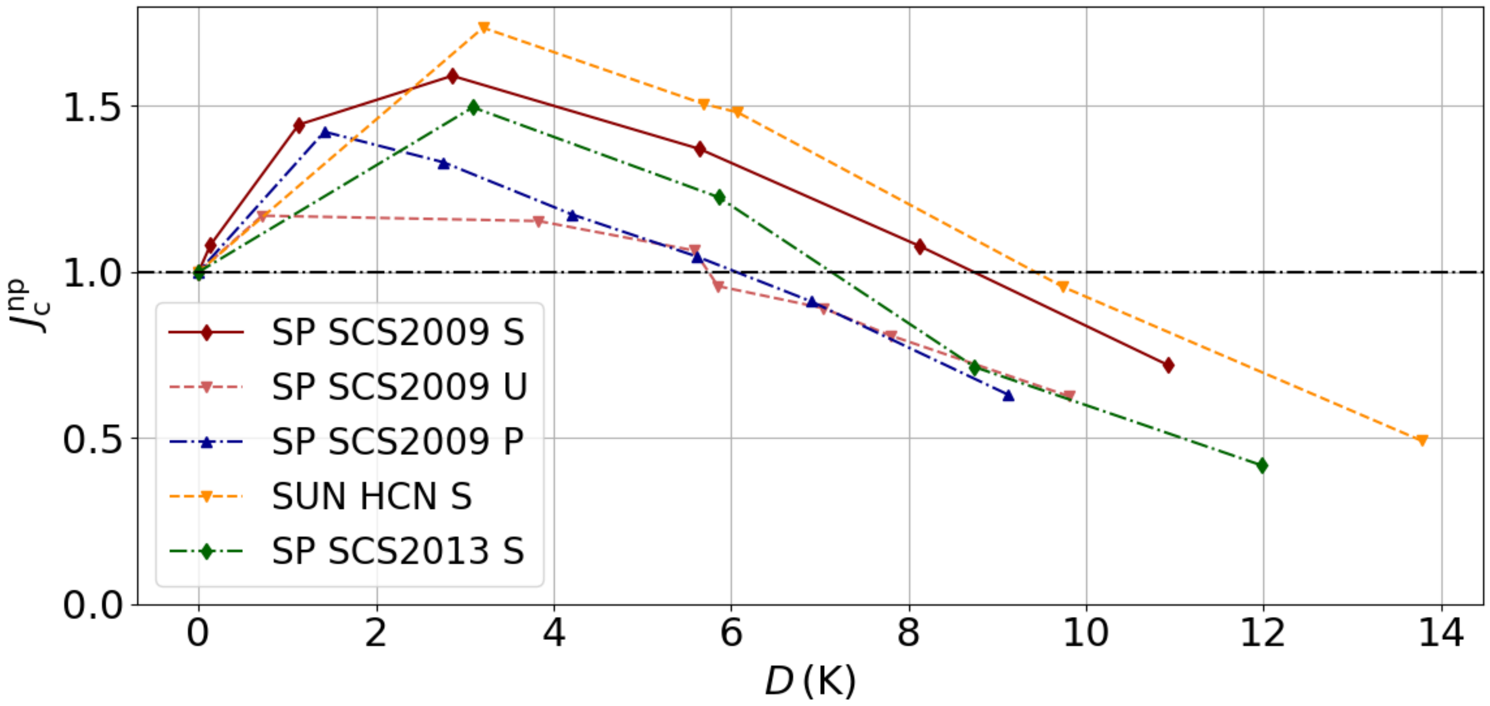
# Influence of thermal neutrons - $J_c$



**Uniform degradation**

- Different defect densities
- Parallel degrading branch
- Specific defects origin of degradation?
- Accumulation in all irradiation techniques?

# Fluence dependence of critical current



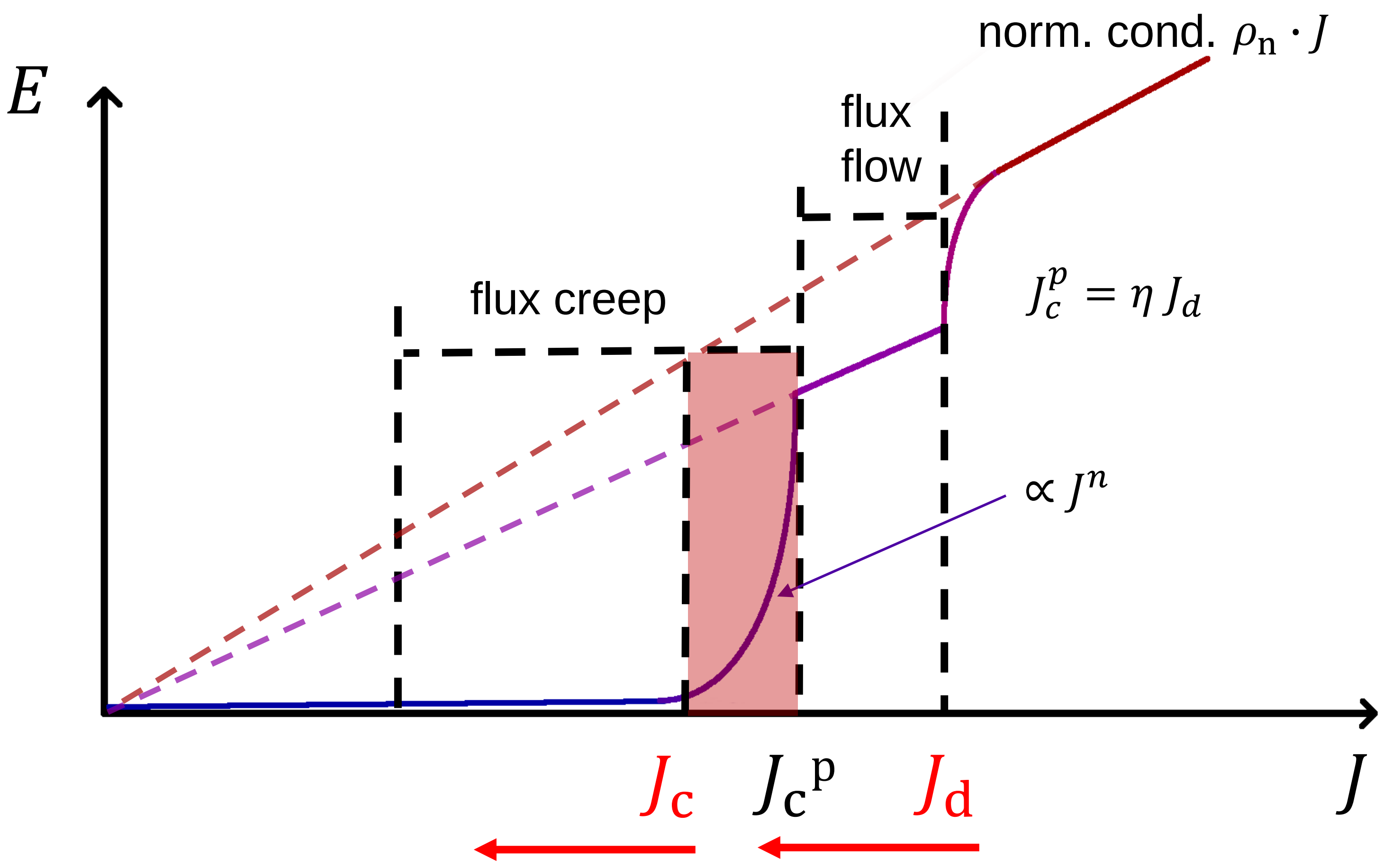
Disorder parameter

$$\boldsymbol{D = -\Delta T_c}$$

measure for scattering

| | |
|---|---|
| $\Phi_\mathrm{f}$ | Fast neutron fluence shielded |
| $\Phi_\mathrm{f}^\mathrm{therm,\,Gd}$ | Fast neutron fluence unshielded |
| $\Phi_\mathrm{p}$ | Proton Fluence |

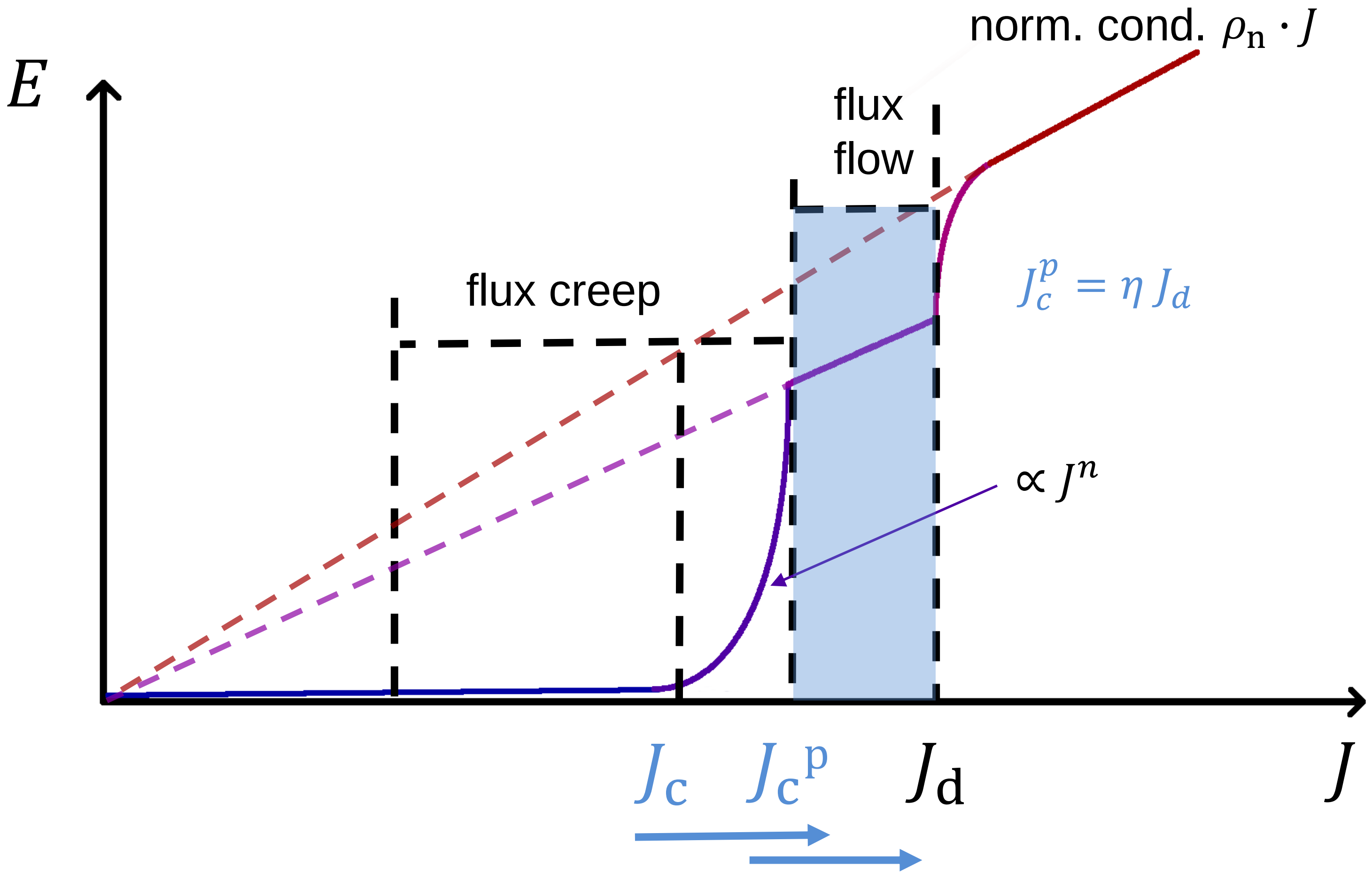
# Sketch of resistive transition



## Degrading scattering

- $T_\mathrm{c}$ decreases
- $J_d$ decreases
- *n*-value decreases

# Sketch of resistive transition



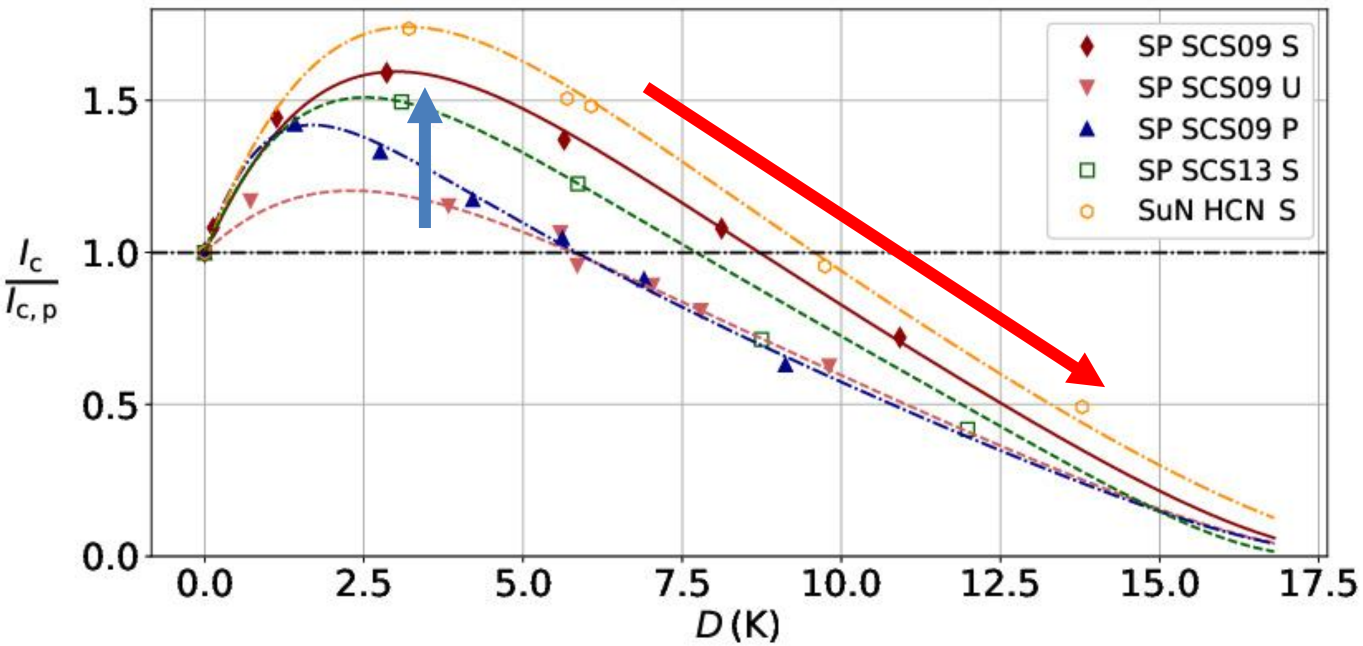
## Degrading scattering

- $T_\mathrm{c}$ decreases
- $J_d$ decreases
- *n*-value decreases

## Enhancing pinning

- $J_c = \eta J_d$
- $\eta$ increases
  (pinning efficiency)

# Modelling



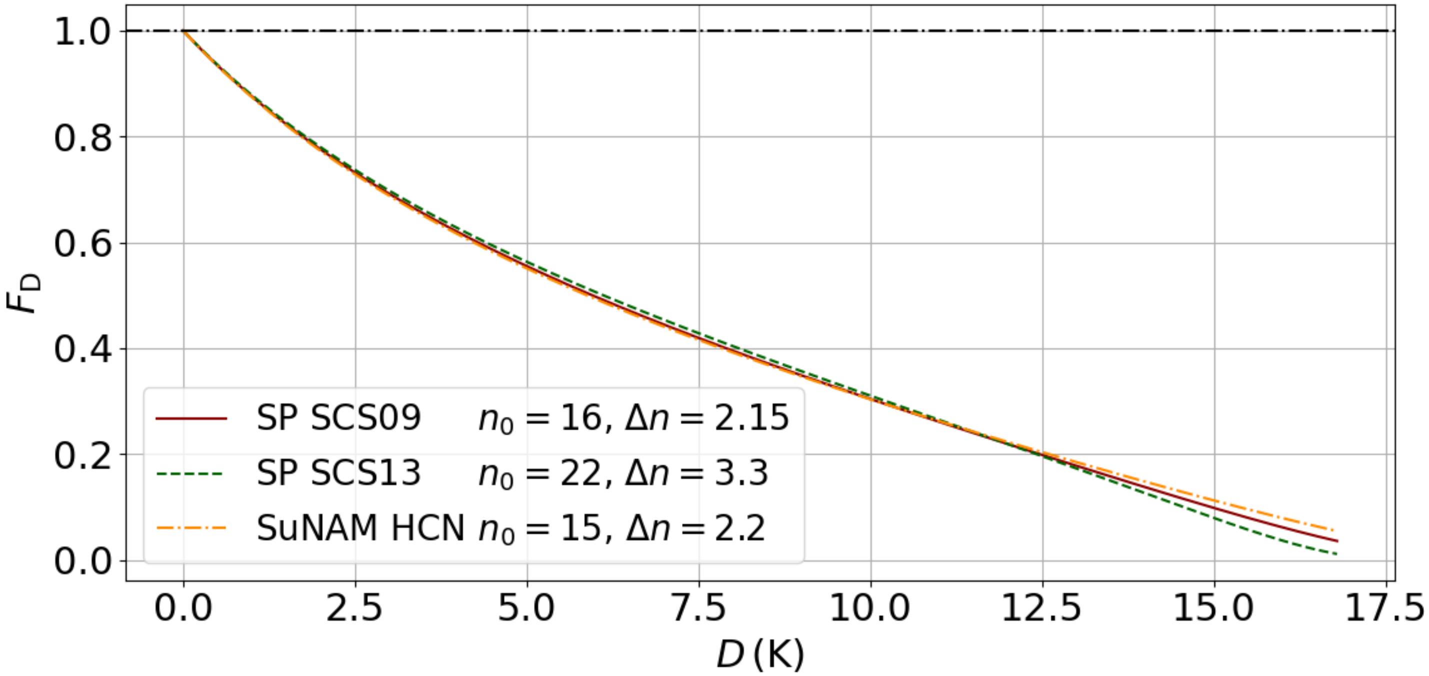
$$\frac{I_c}{I_{c,p}} = \sqrt{\frac{(\alpha_p + 1) t_c^3}{\alpha_p \left(1 - K_\rho (1 - t_c)\right) + t_c}} \left(\frac{E_{crit}}{\sqrt{\phi_0 B \nu_0}}\right)^{\frac{1}{n} - \frac{1}{n_p}} \frac{\eta_{max}}{\eta^0} \tanh\left(\frac{D}{D_{\eta_{max}}} (\pi - A) + A\right) \qquad t_c = \frac{T_c}{T_{c,p}}$$

Degrading - $F_D$

Enhancing - $\eta$

# Degradation function

$$\frac{I_\mathrm{c}}{I_\mathrm{c,p}} = \sqrt{\frac{(\alpha_\mathrm{p}+1)t_c^3}{\alpha_\mathrm{p}\left(1-K_\rho(1-t_\mathrm{c})\right)+t_\mathrm{c}}}\left(\frac{E_\mathrm{crit}}{\sqrt{\phi_0 B \nu_0}}\right)^{\frac{1}{n}-\frac{1}{n_\mathrm{p}}} \frac{\eta_\mathrm{max}}{\eta^0}\tanh\left(\frac{D}{D_{\eta_\mathrm{max}}}(\pi-A)+A\right) \qquad t_\mathrm{c} = \frac{T_\mathrm{c}}{T_\mathrm{c,p}}$$



Attempt frequency $\nu_0 = 2.5 \times 10^7\ \mathrm{Hz}$

Electric. field criterion $E_\mathrm{c} = 1\ \mu\mathrm{V\ cm}$

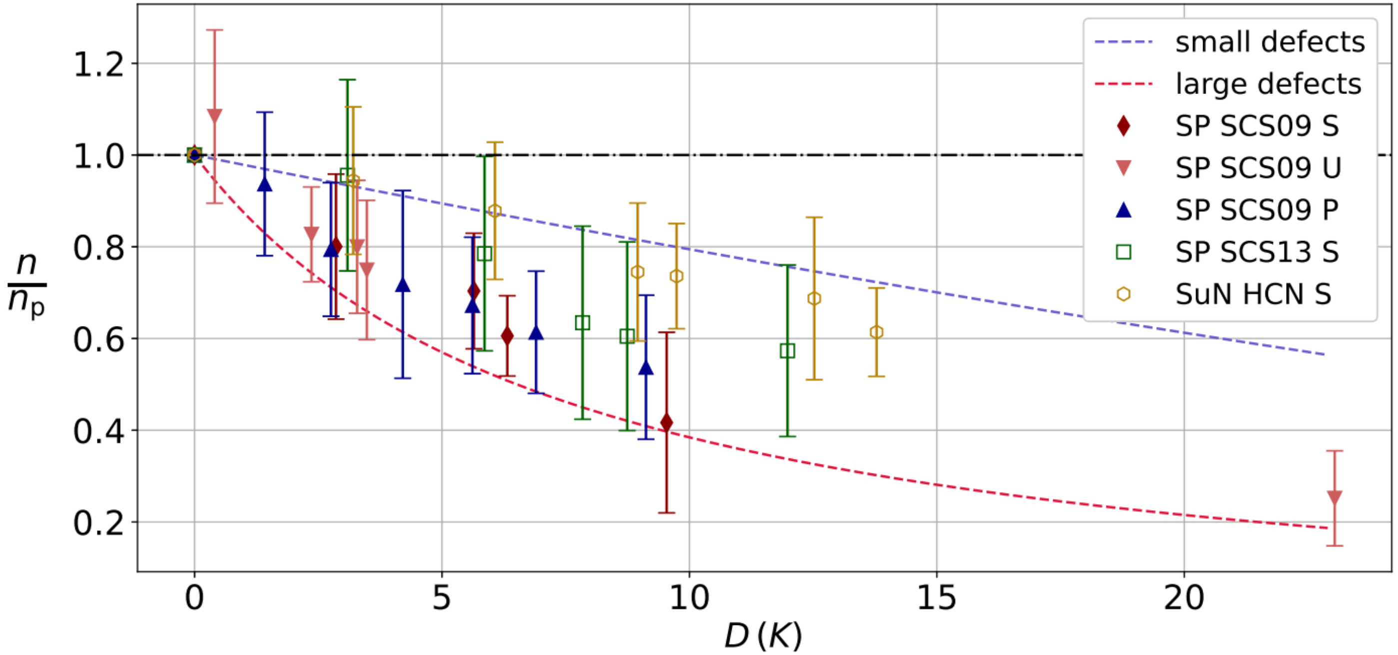
$\alpha = \frac{\xi_0}{l} = 3 \qquad K_\rho = \frac{T_\mathrm{c,p}}{\rho_\mathrm{n,p}}\frac{\partial \rho_\mathrm{n}}{\partial T_\mathrm{c}} = -16.5$

Experimetal $n = n_0 - \Delta n D$

- Universal
- All parameters fairly easily accessible
- Curve hard to measure directly due to pinning

# Degradation function

$$\frac{I_\mathrm{c}}{I_\mathrm{c,p}} = \sqrt{\frac{(\alpha_\mathrm{p}+1)t_c^3}{\alpha_\mathrm{p}\left(1-K_\rho(1-t_\mathrm{c})\right)+t_\mathrm{c}}}\left(\frac{E_\mathrm{crit}}{\sqrt{\phi_0 B \nu_0}}\right)^{\frac{1}{n}-\frac{1}{n_\mathrm{p}}} \frac{\eta_\mathrm{max}}{\eta^0}\tanh\left(\frac{D}{D_{\eta_\mathrm{max}}}(\pi-A)+A\right) \qquad t_\mathrm{c}=\frac{T_\mathrm{c}}{T_\mathrm{c,p}}$$

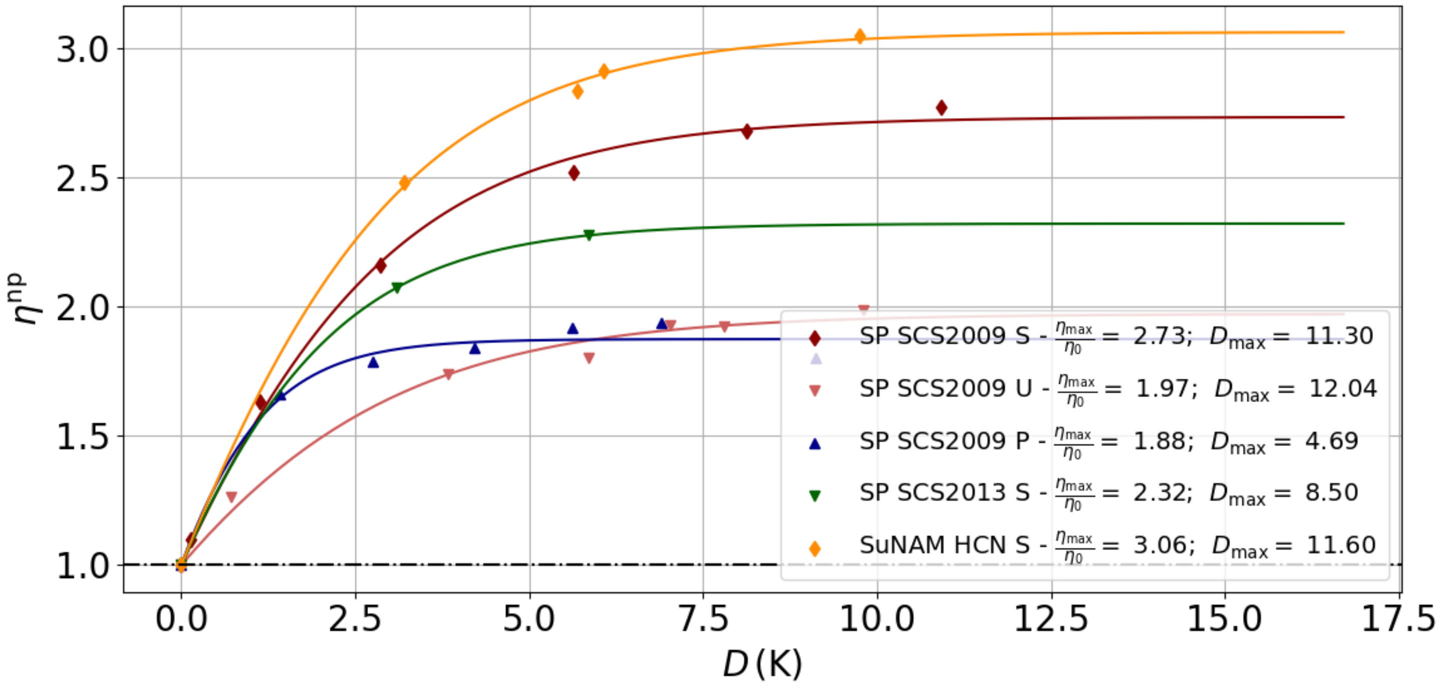


- $J \propto E^n$
- Linear fit to experimental data
  $n = n_0 - \Delta n D$
- Smoothening of scattered data
- Envelope of expectations for large and small defects

# Pinning enhancement

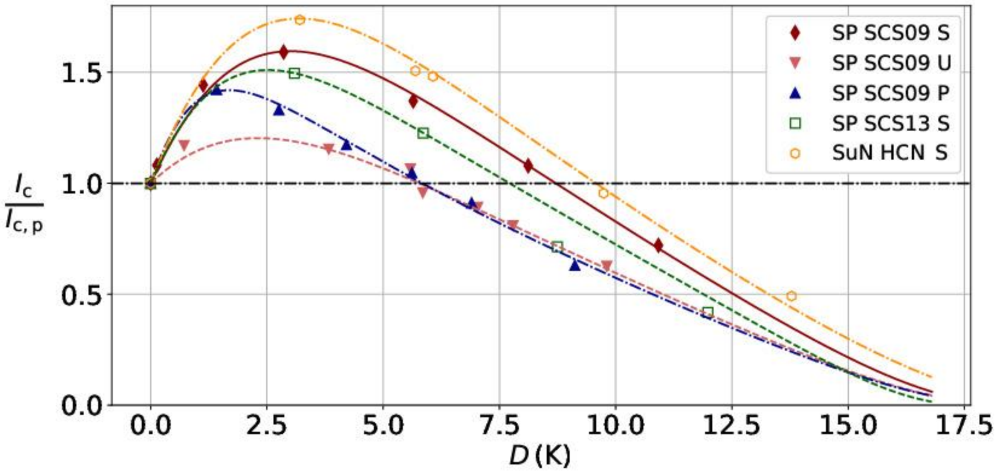
$$\frac{I_{\mathrm{c}}}{I_{\mathrm{c,p}}} = \sqrt{\frac{(\alpha_{\mathrm{p}}+1)t_c^3}{\alpha_{\mathrm{p}}\left(1-K_\rho(1-t_{\mathrm{c}})\right)+t_{\mathrm{c}}}}\left(\frac{E_{\mathrm{crit}}}{\sqrt{\phi_0 B \nu_0}}\right)^{\frac{1}{n}-\frac{1}{n_{\mathrm{p}}}} \frac{\eta_{\mathrm{max}}}{\eta^0}\tanh\left(\frac{D}{D_{\eta_{\mathrm{max}}}}(\pi - A) + A\right) \qquad t_{\mathrm{c}} = \frac{T_{\mathrm{c}}}{T_{\mathrm{c,p}}}$$

$$A = \tanh^{-1}\frac{\eta^0}{\eta_{\mathrm{max}}}$$



- tanh was chosen for the good correspondence to the data
- Pinning efficiency can not increase above ~ 0.3

# Conclusions



$$\frac{I_{\mathrm{c}}}{I_{\mathrm{c,p}}} = \sqrt{\frac{(\alpha_{\mathrm{p}}+1)t_c^3}{\alpha_{\mathrm{p}}\left(1-K_\rho(1-t_{\mathrm{c}})\right)+t_{\mathrm{c}}}}\left(\frac{E_{\mathrm{crit}}}{\sqrt{\phi_0 B \nu_0}}\right)^{\frac{1}{n}-\frac{1}{n_{\mathrm{p}}}} \frac{\eta_{\max}}{\eta^0}\tanh\left(\frac{D}{D_{\eta_{\max}}}(\pi-A)+A\right)$$

**Degradation** is nearly universal and driven by the loss of superfluid density.

- Physical origin: Pair breaking scattering on small defects
- Relevant parameter: Scattering rate $\tau^{-1}$ (numerical modelling?)
- Experimental manifestation: Decrease of transition temperature ($D$), normal state resistivity ($\rho_{\mathrm{n}}$)

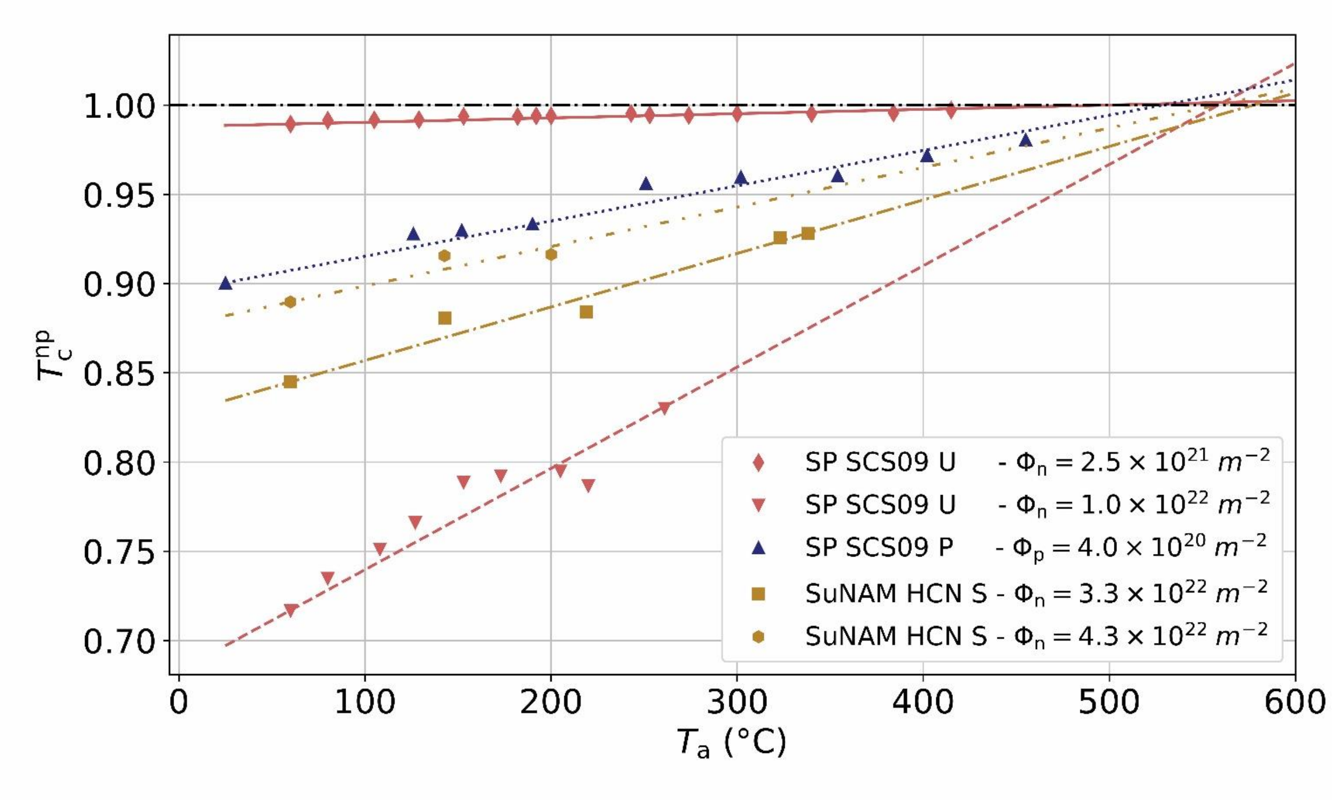
**Increase** in critical current results from improved pinning

- Non-universal, depending on initial defect structure and type of radiation.
- Physical origin: Flux pinning by large defects
- Relevant parameter: pinning efficiency $\eta$ (TDGL modelling?)
- Experimental benchmarking: Saturation value of $\eta$ (suitable proxies for neutrons?)

# Annealing

# Thermal stability of small vs large defects

annealing

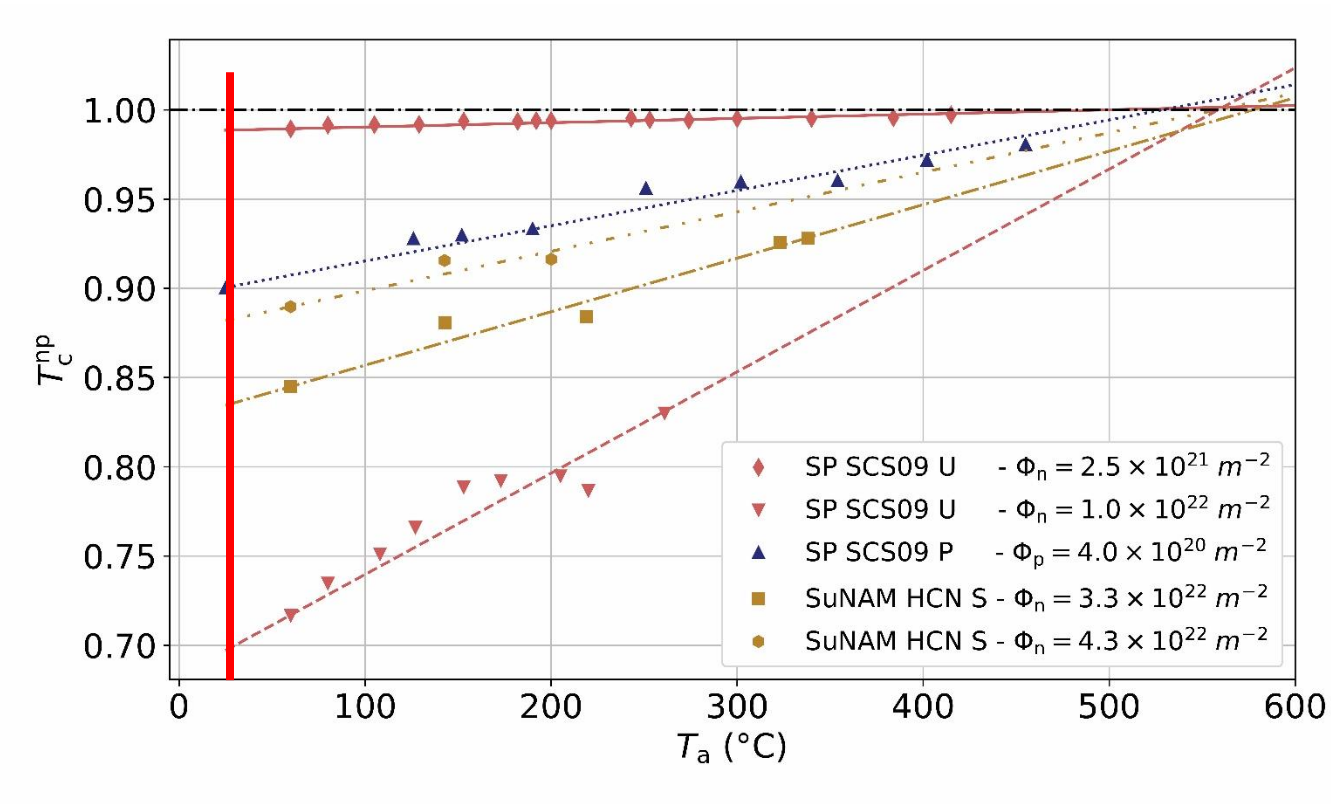


- $T_c$ regenerates "linearly" with $T_a$
- All neutron and proton irradiated samples anneal to same point
- Annealing defects have same/similar distribution and activation barrier
- $n_{therm}$, $n_{fast}$ & $p^+$ irradiated samples

Samples annealed in pure $O_2$ atmosphere at 1 atm

# Thermal stability of small vs large defects

annealing

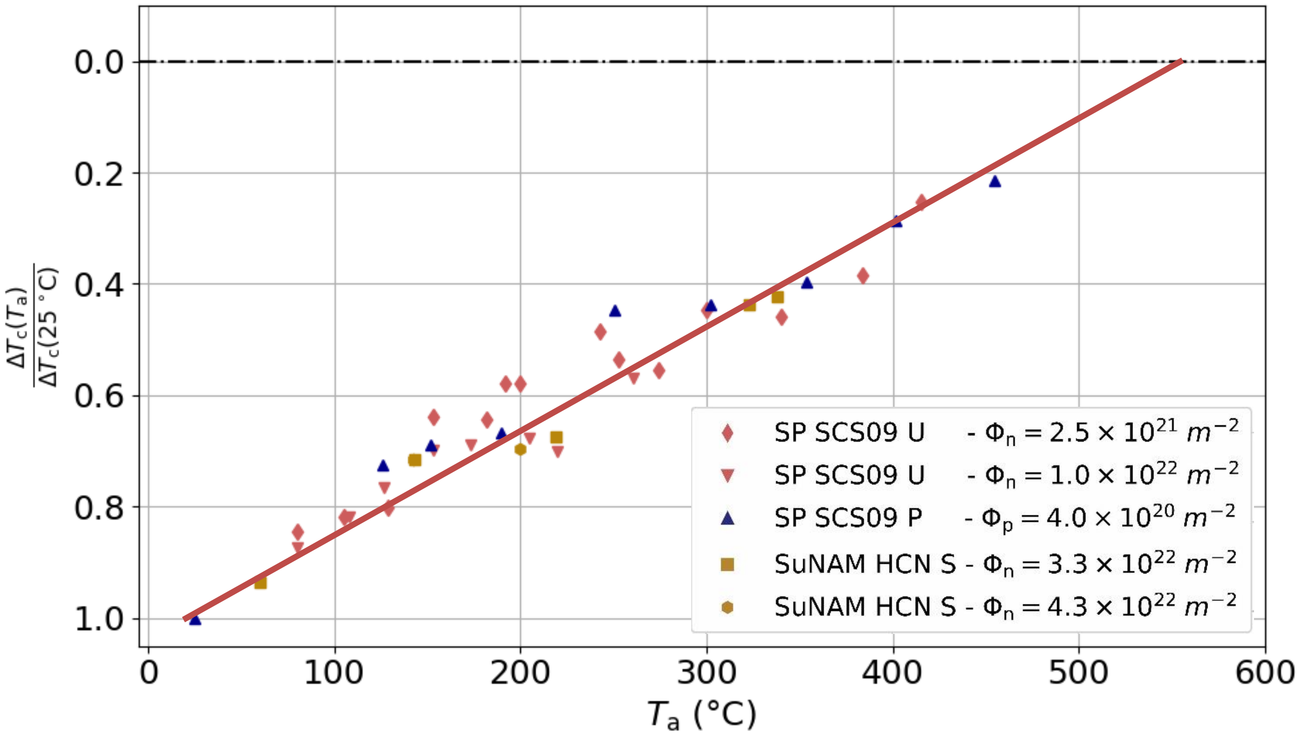


Normalizing $\Delta T_c(T_a)$ to $\Delta T_c(T_a = 25\ °C)$

- $T_c$ regenerates “linearly” with $T_a$
- All neutron and proton irradiated samples anneal to same point
- Annealing defects have same/similar distribution and activation barrier
- $n_{therm}$, $n_{fast}$ & $p^+$ irradiated samples

# Thermal stability of small vs large defects

annealing

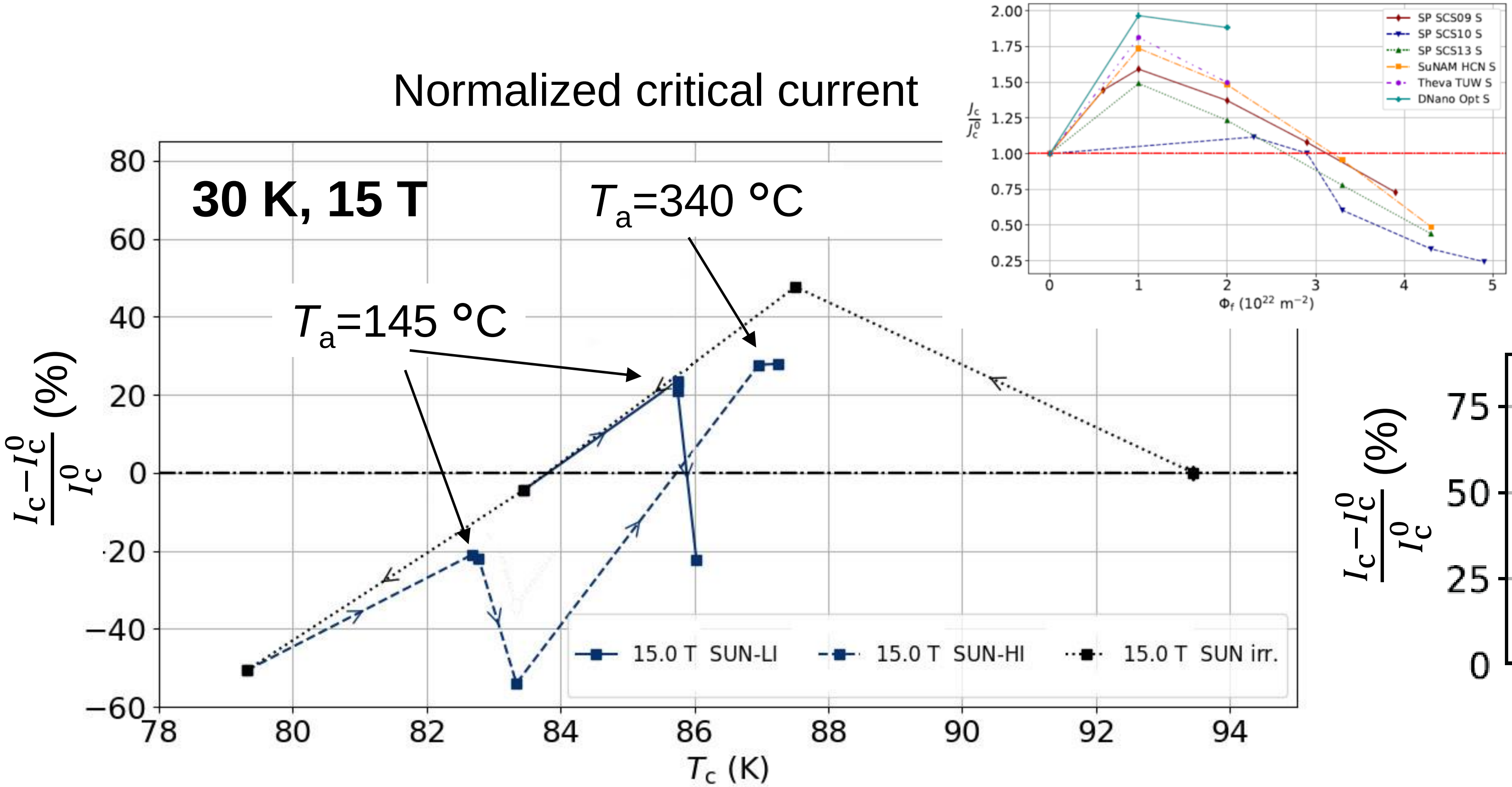


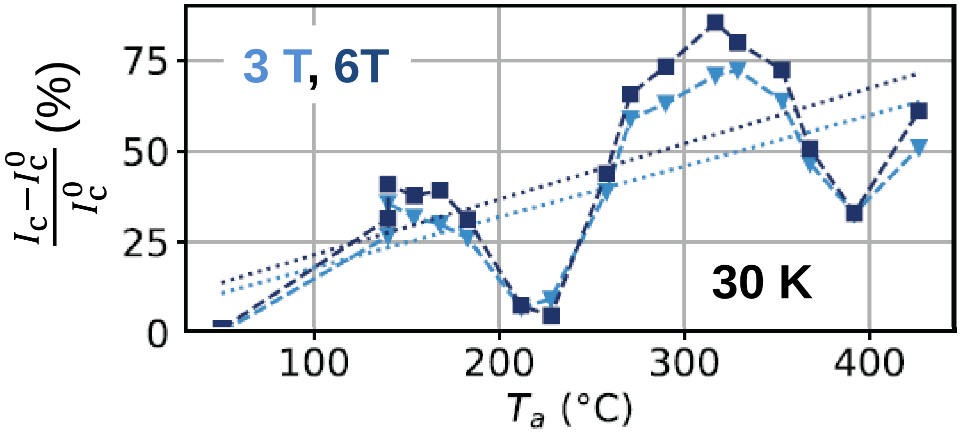
Normalizing $\Delta T_c(T_a)$ to $\Delta T_c(T_a = 25\ ^\circ C)$

- $T_c$ regenerates “linearly” with $T_a$
- All neutron and proton irradiated samples anneal to same point
- Annealing defects have same/similar distribution and activation barrier
- $n_{therm}$, $n_{fast}$ & $p^+$ irradiated samples

# Annealing: $I_c$

Normalized critical current





- Recovery is non-monotonous, although with a linear trend.
- Degraded samples was recovered above its initial value.
- Optimum annealing protocol to be derived.

R. Unterrainer et al., SuST 35 (2022) 04LT01

# Contribution to the workflow

- Fast neutron irradiation is a benchmarking experiment
  - Among the closest avaible neutron sprectrum
- Checks the reability of Tasks 2) *Defect Structure* and 3) *Flux Pinning and Superfluid Density.*
  - Task 2) provides the input parameters ($\tau^{-1}$, defect density and size) for Task 3)
  - Task 3) predicts changes of $I_c$ and $T_c$.
- Comparison with experimental values.