Measuring radiation damage dynamics by pulsed ion beam irradiation

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Sergei O. Kucheyev

Lawrence Livermore National Laboratory

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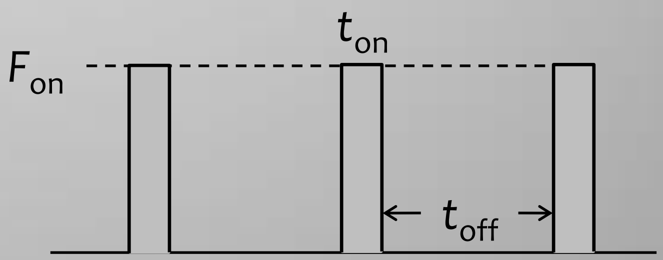
LLNL-PRES-700857

Outline

- Team
- Importance of radiation dynamics
- Pulsed-beam method to study defect dynamics
  - Time constant τ
  - Diffusion length L_d
- Results for SiC and Si
- Implications



Current team

- Sergei Kucheyev (LLNL) – PI
- L. Bimo Bayu Aji (LLNL) – postdoc

- Joseph Wallace (LLNL & TAMU) – 100% LGSP support
- Lin Shao (TAMU) – graduate adviser of Joseph Wallace

- Swanee Shin (LLNL) – TEM

Scientific objectives and relevance

Objectives

Our objective is to develop a novel ***pulsed-ion-beam method*** to access the dynamic regime of radiation damage formation in nuclear materials and to measure

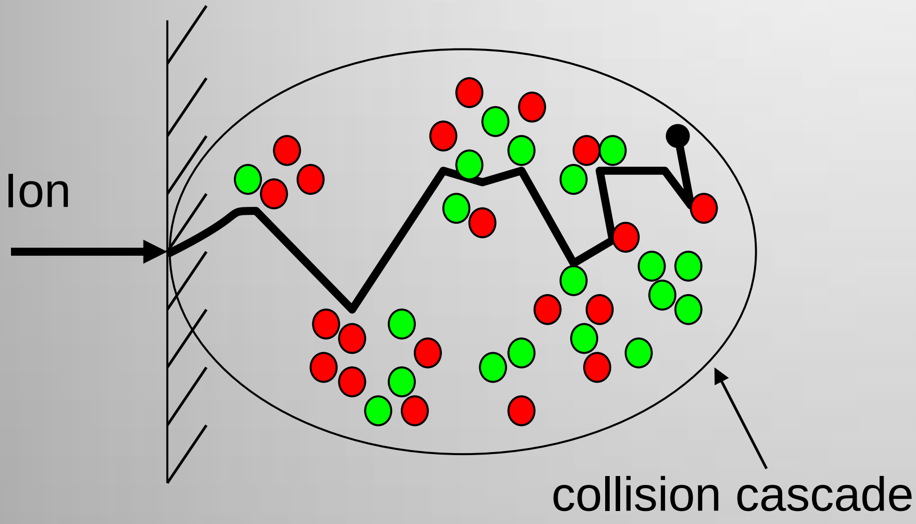
- defect interaction time constants,
- defect diffusion lengths,
- activation energies of defect interaction processes.

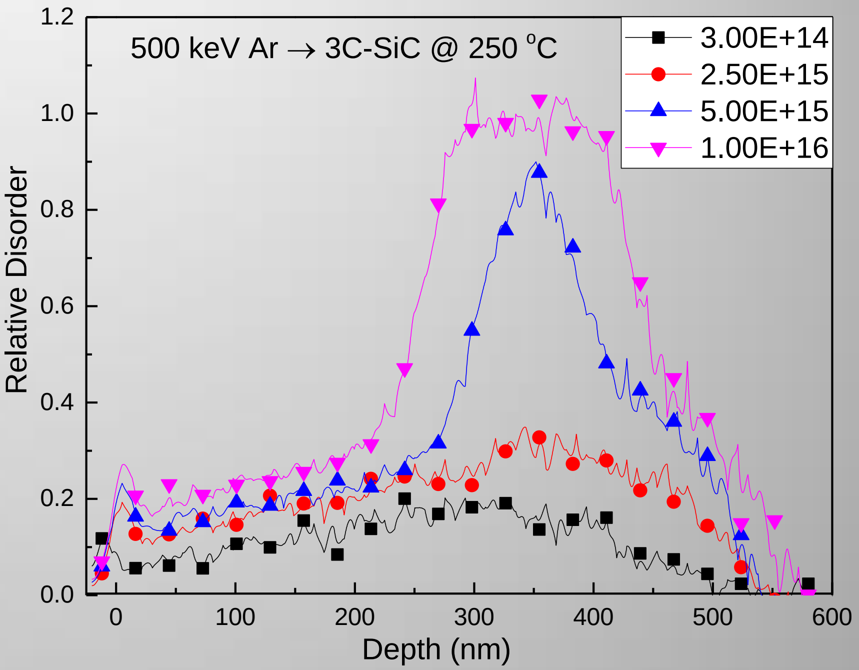
Relevance

This project could establish the pulsed-ion-beam method as the primary approach to study defect interaction dynamics in nuclear materials.

- Development of new reactor materials.
- Extending laboratory findings to nuclear material lifetimes.
- Data on defect dynamics for building realistic radiation damage models.
- Understanding limitations of using rastered ion beams to simulate neutron-induced radiation damage in nuclear materials.

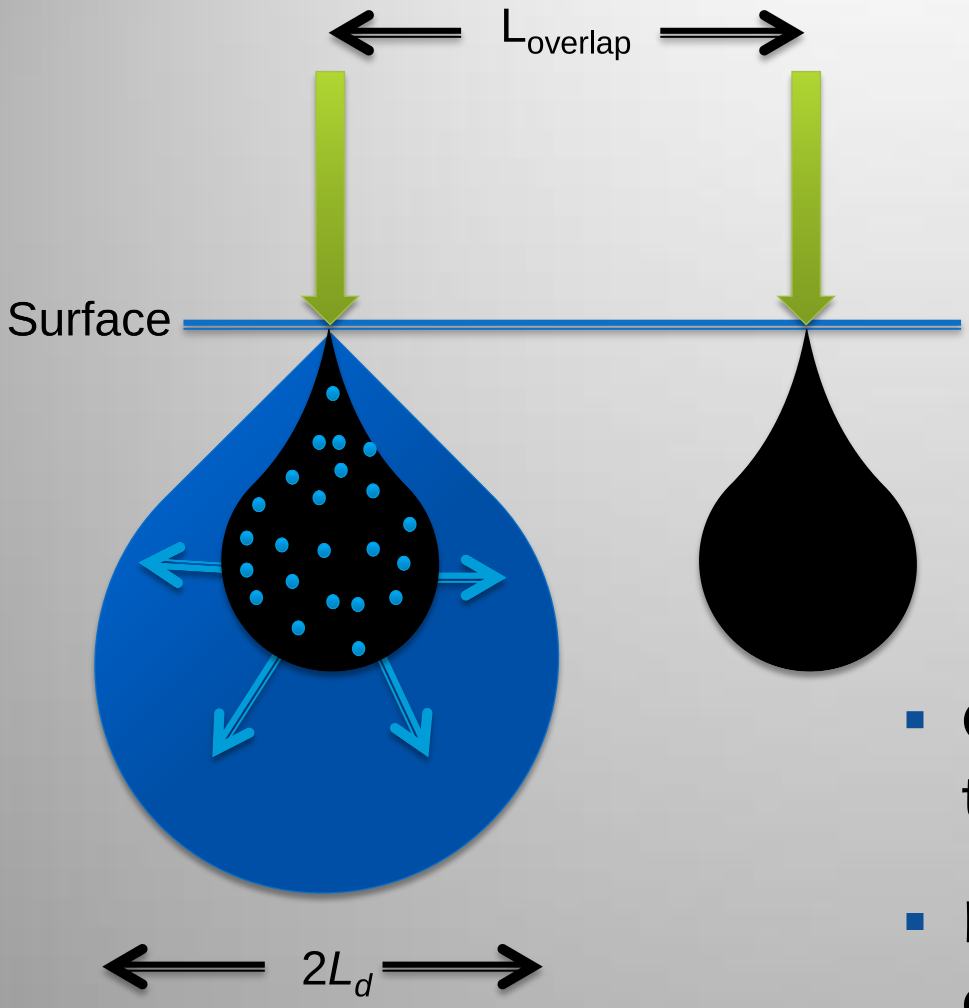
We lack predictive capabilities to describe radiation damage in nuclear materials

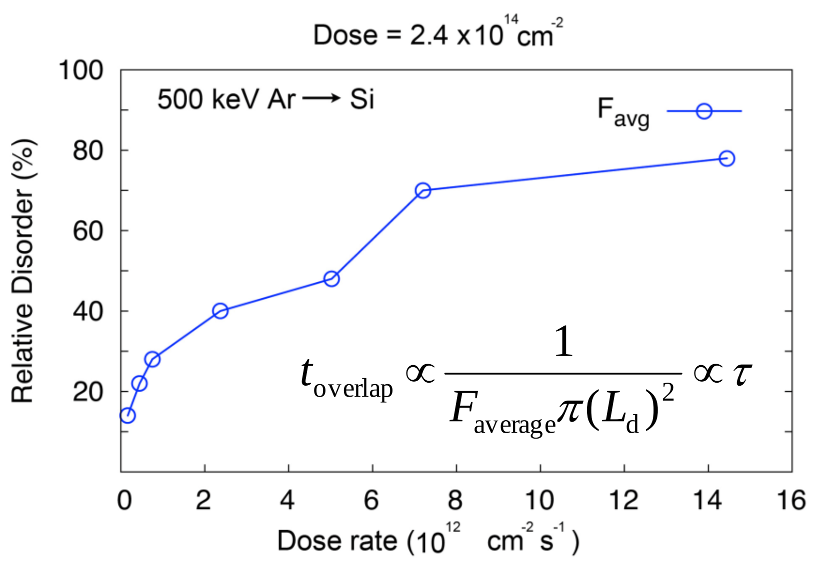




The goal of this project is to develop innovative defect characterization methodology allowing unprecedented access to the dynamic regime of damage formation.

Radiation damage is dynamic phenomenon

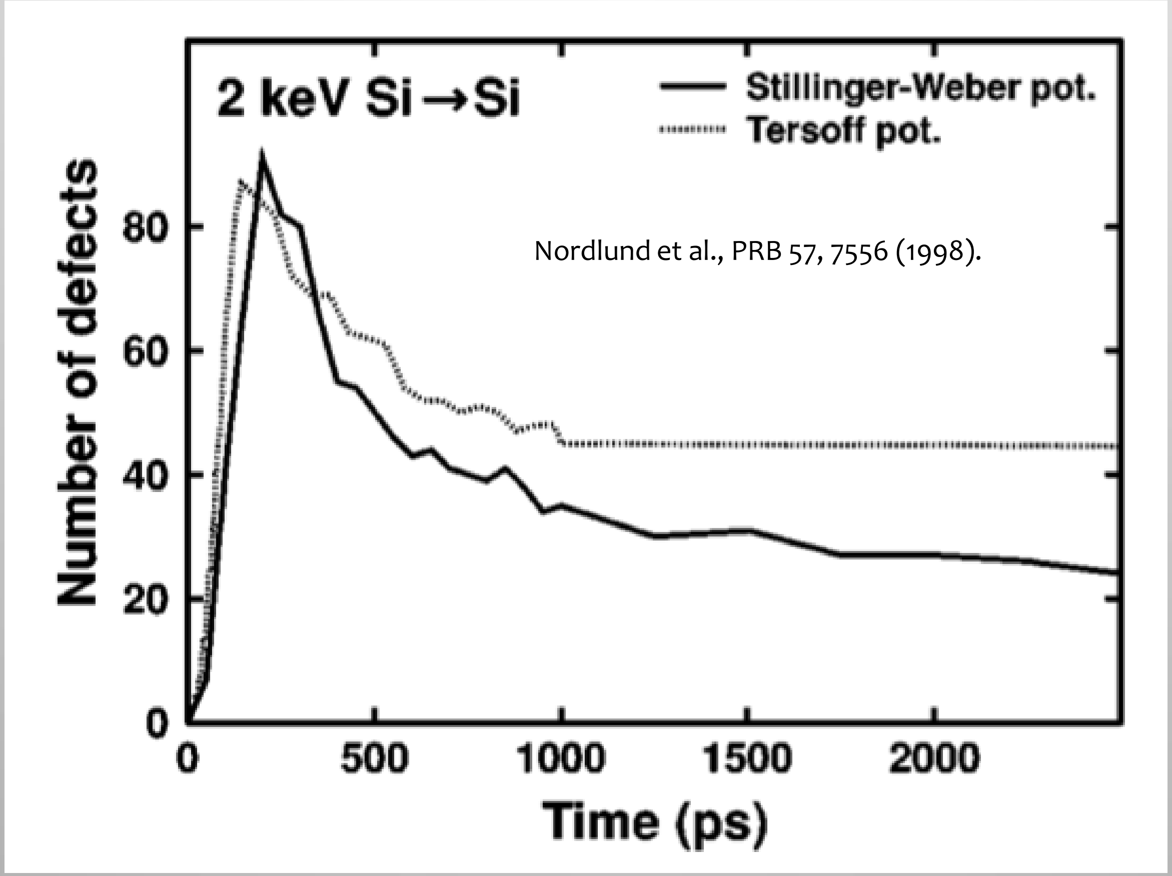




$$t_{\text{overlap}} \propto \frac{1}{F_{\text{average}}\pi(L_{\text{d}})^2} \propto \tau$$

- Cascade formation and thermalization (<1 ps).
- Dynamic annealing: defects diffuse distance L_d over time τ.

Previous estimates of τ (Si @ RT): Molecular dynamics



Nordlund et al., PRB 57, 7556 (1998).

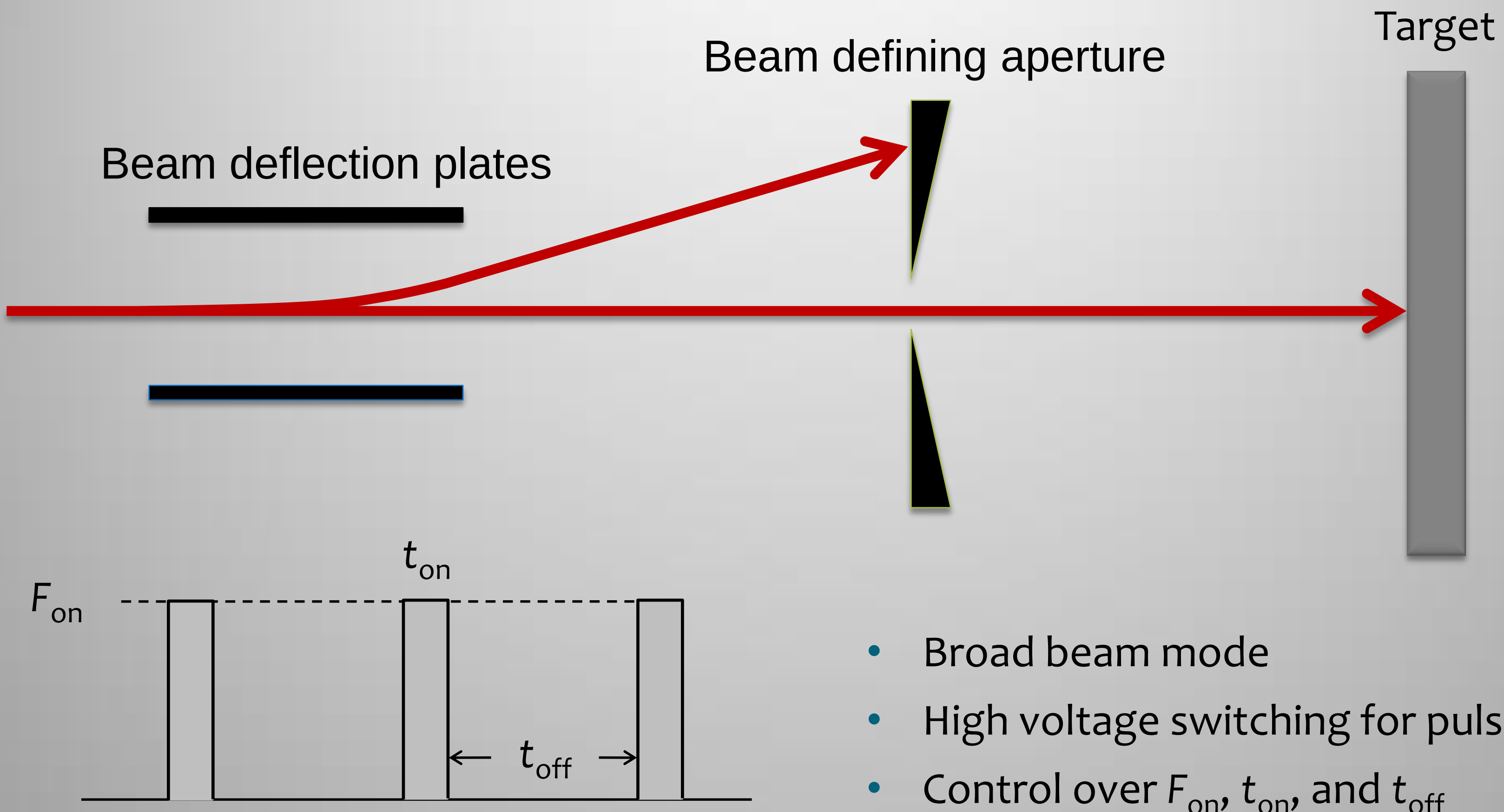
$\tau \sim 1$ ns

Large discrepancy in τ and L_d even for Si@RT

- τ
  - MD: ~ 10^{-10} s [1]
  - Flux effect: ~ 10^{-2} - 10^{+2} s [2-3]
- L_d
  - Flux effect: ~10-50 nm [3-4]
  - DLTS/SRP/PL: ~50-2000 nm [5-7]
  - In-situ TEM: ~20-30 nm [8-9]
- Novel approaches to study defect dynamics are needed.

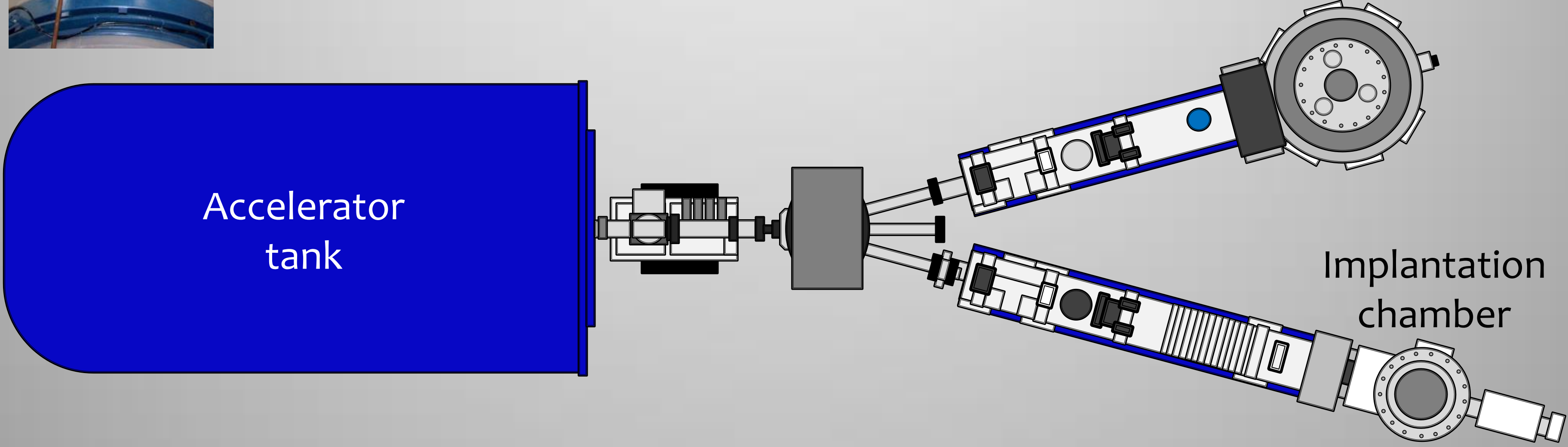
[1] Nordlund et al., PRB 57, 7556 (1998).
[2] Posselt et al., APL 79, 1444 (2001).
[3] Titov et al., PRB 73, 064111 (2006).
[4] Titov et al., NIMB 212, 169 (2003).
[5] Benton et al., JVST B 10, 540 (1992).
[6] Larsen et al., PRL 76, 1493 (1996).
[7] Deenapanray, APL 80, 1577 (2002).
[8] Ruault et al., NIM 209, 351 (1983).
[9] Lulli et al., PRB 36, 8038 (1987).

Pulsed-beam method



- Broad beam mode
- High voltage switching for pulsing
- Control over F_{on}, t_{on}, and t_{off}

4 MV Accelerator Facility @ LLNL



Accelerator tank

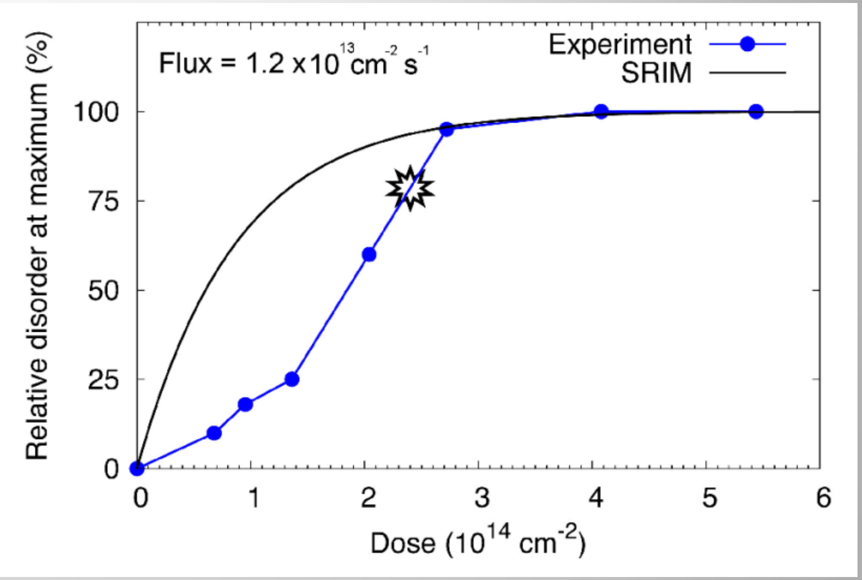
Analysis chamber

Implantation chamber

Pulsed beam method

1. Continuous beam: Damage buildup

    - t_{off} = 0 ms
    - Flux = *const*
    - Dose = *variable*

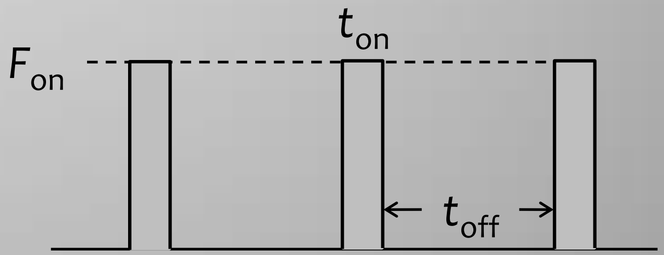


2. Pulsed beam: t_{off} dependence $\Rightarrow$ τ

    - t_{off} = *variable*

3. Pulsed beam: t_{on} dependence $\Rightarrow$ L_d

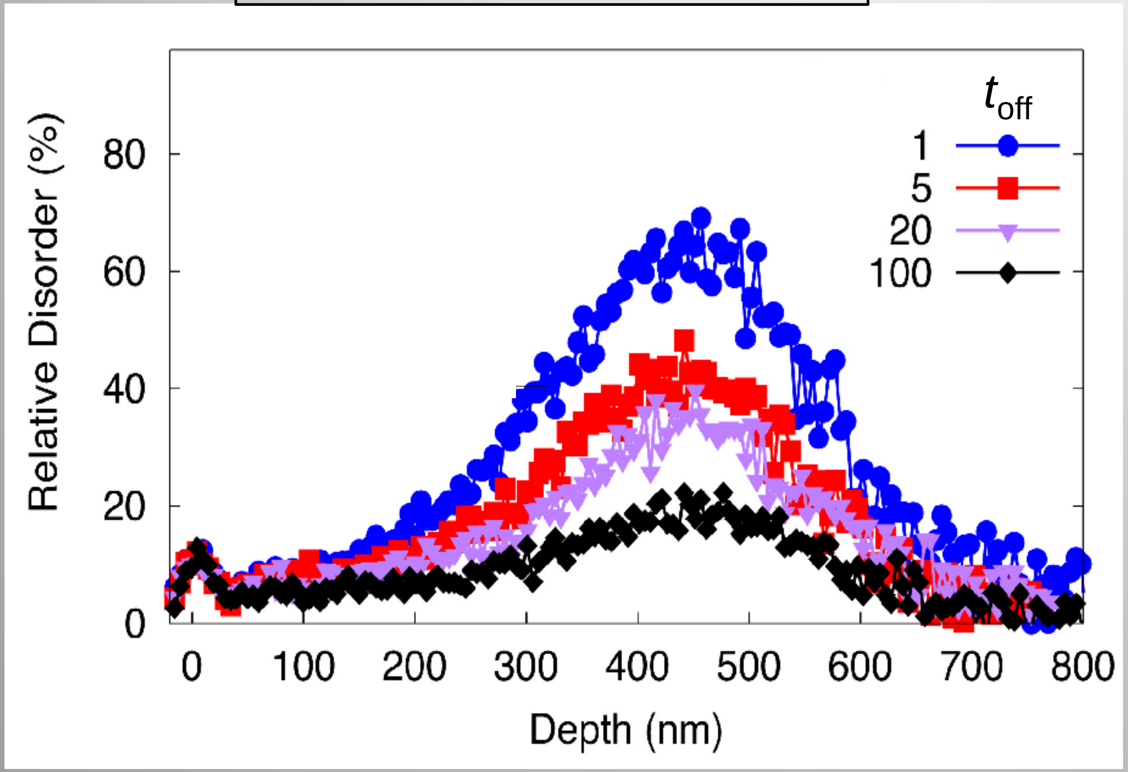
    - t_{on} = *variable*
    - $t_{off} >> \tau$

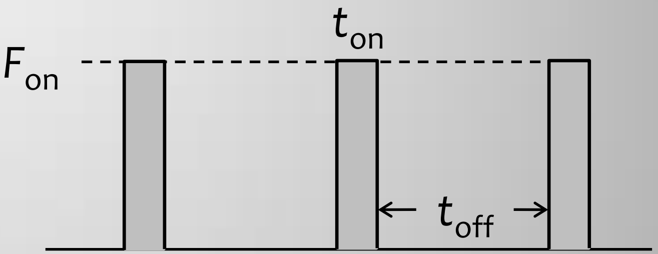


4. Pulsed beam to study defect physics: material, temperature, ion mass, and t_{on} dependencies and correlation with simulations.

Measurement of τ: damage dependence on t_{off}

500 keV ^{40}Ar → FZ Si @RT

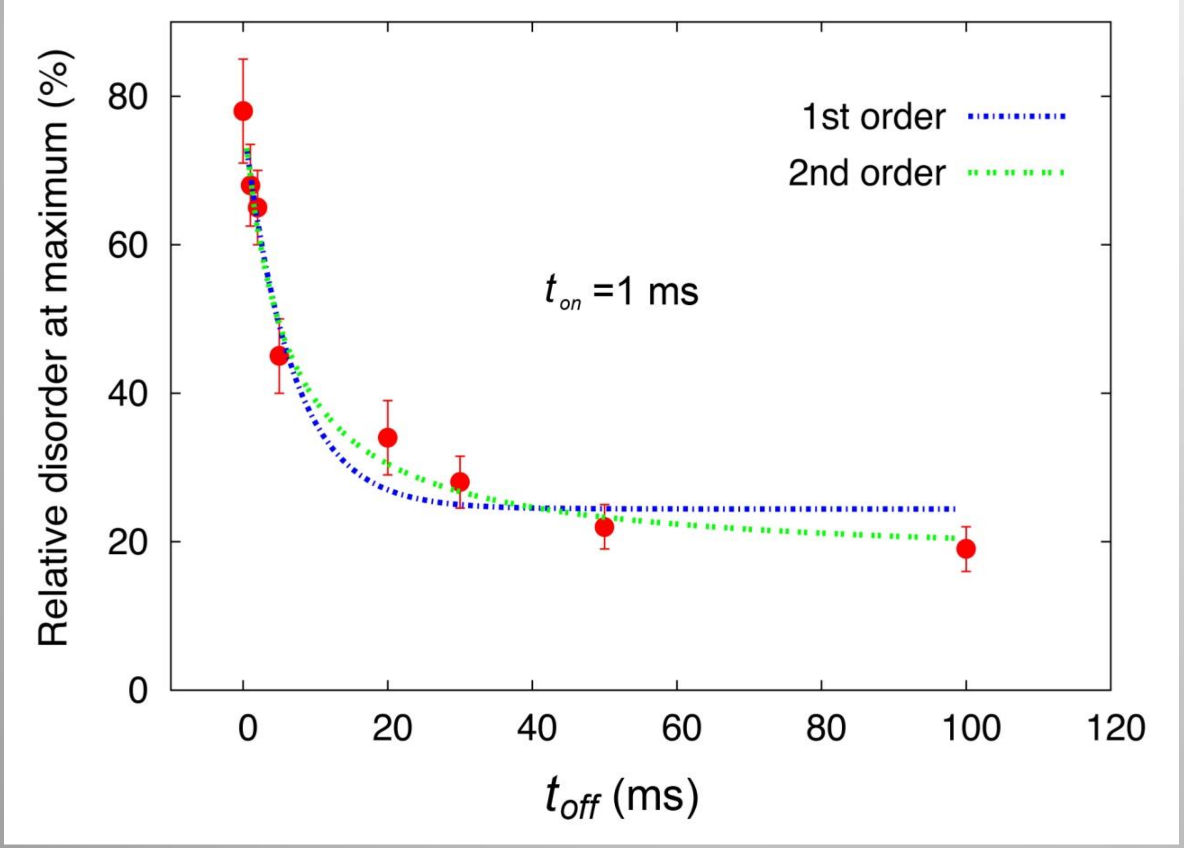


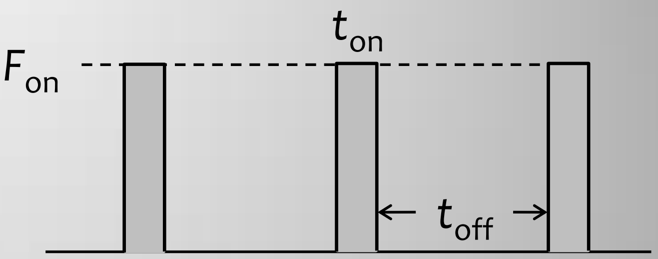


- Dose = 2.4×10^{14} cm^{-2}
- $F_{on} = 1.2\times10^{13}$ cm^{-2} s^{-1}
- t_{on} = 1 ms (~ 20,000 pulses)
- Less bulk damage for longer t_{off}
- Surface disorder is independent of t_{off}

Phys. Rev. Lett. 109, 095502 (2012).

For Si @RT (500 keV Ar): τ ~ 6 ms





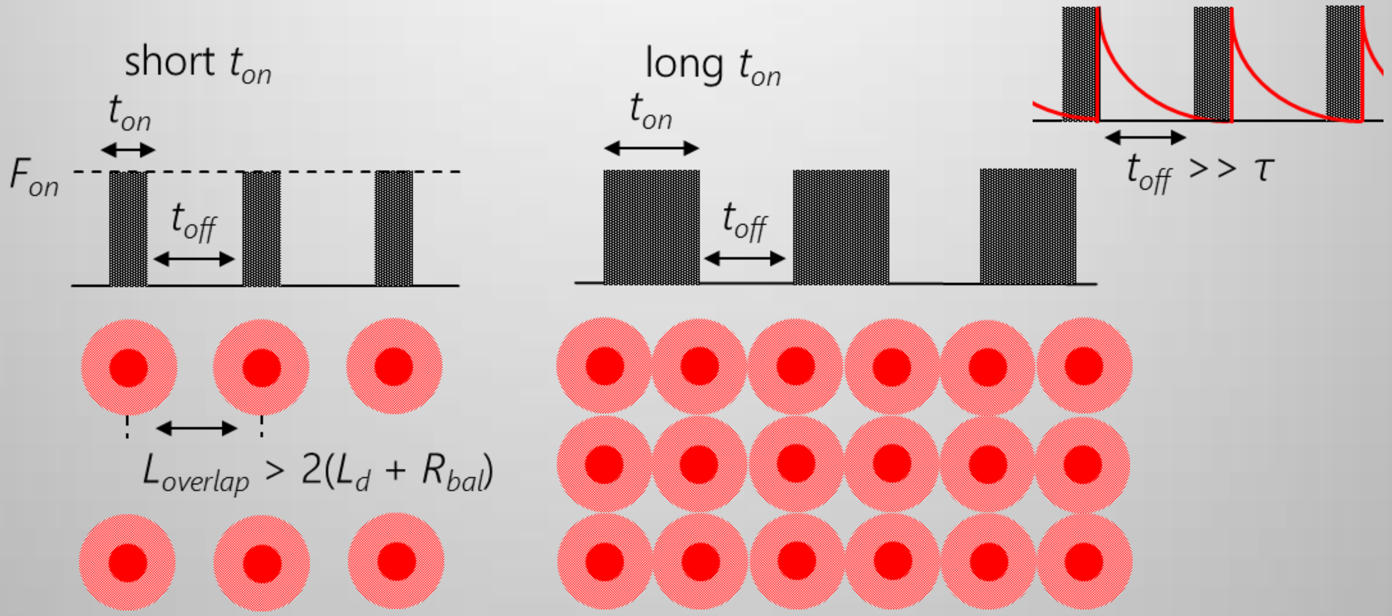
- 2nd order kinetics

$$\frac{\partial}{\partial t} n_d \propto n_d^2 \qquad n_d = n_\infty + \frac{n_0 - n_\infty}{1 + \frac{t_{off}}{\tau}}$$

- τ ~ 6 ms

Phys. Rev. Lett. 109, 095502 (2012).

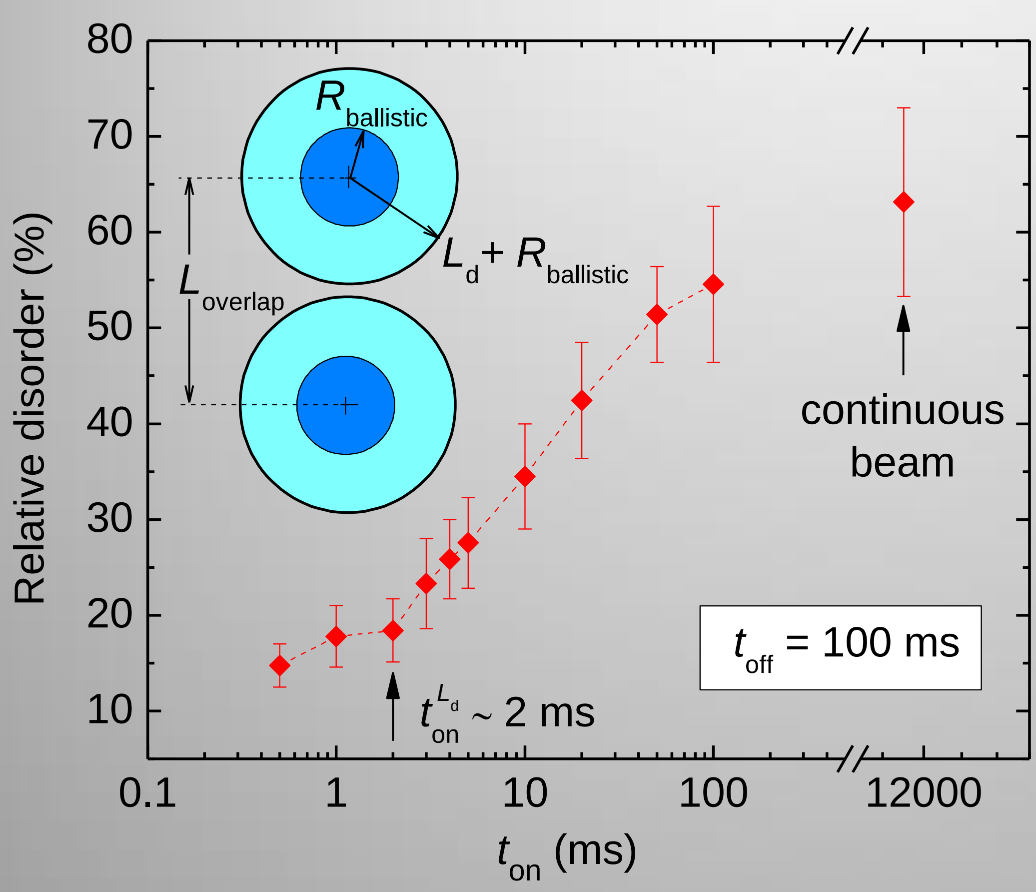
Measurement of L_d :
The dependence of damage on t_{on}

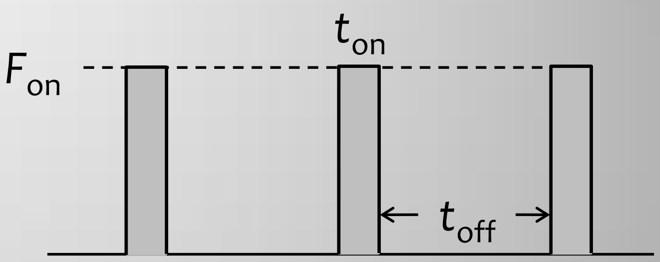


Defects from different cascades **do not** interact

Defects from different cascades interact

For Si @RT (500 keV Ar): $L_d \sim 30$ nm





$$L_{\text{overlap}} \approx \frac{1}{\sqrt{\Phi_{\text{pulse}}}} = \frac{1}{\sqrt{F_{\text{on}} t_{\text{on}}}}$$

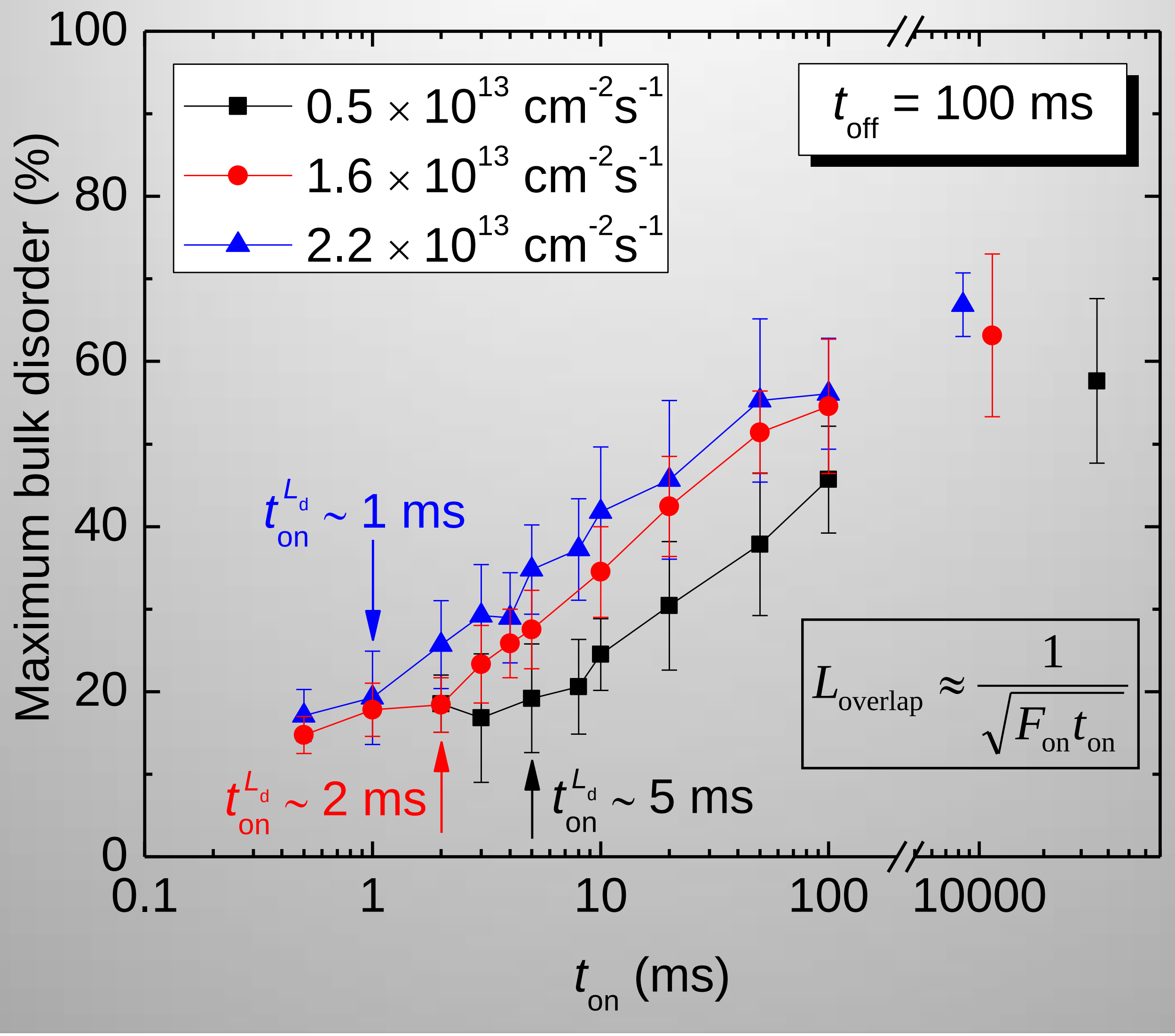
$$L_{\text{overlap}} < 2(L_d + R_{\text{ballistic}}) \approx 2L_d$$

$$t_{\text{on}}^{L_d} = \frac{1}{2\sqrt{F_{\text{on}} t_{\text{on}}^{L_d}}}$$

$$t_{\text{on}}^{L_d} \approx 2\text{ ms} \Rightarrow L_d \approx 30\text{ nm}$$

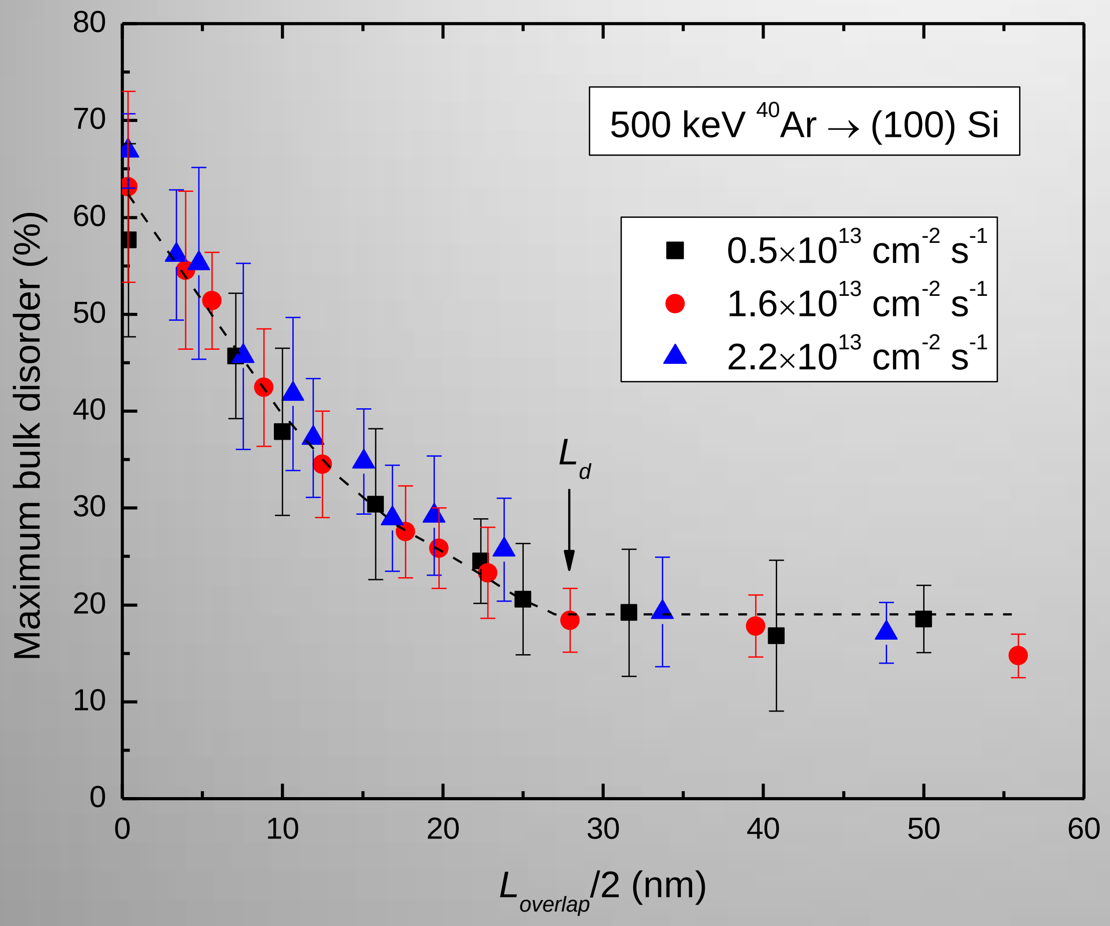
JPCM 25, 162203 (2013).

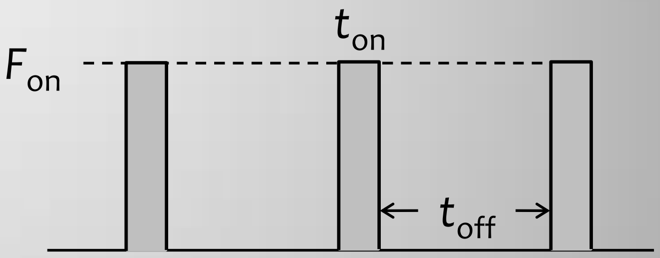
Si@RT (500 keV Ar): $L_d \sim 30$ nm



JAP 118, 135709 (2015)

Si@RT (500 keV Ar): $L_d \sim 30$ nm





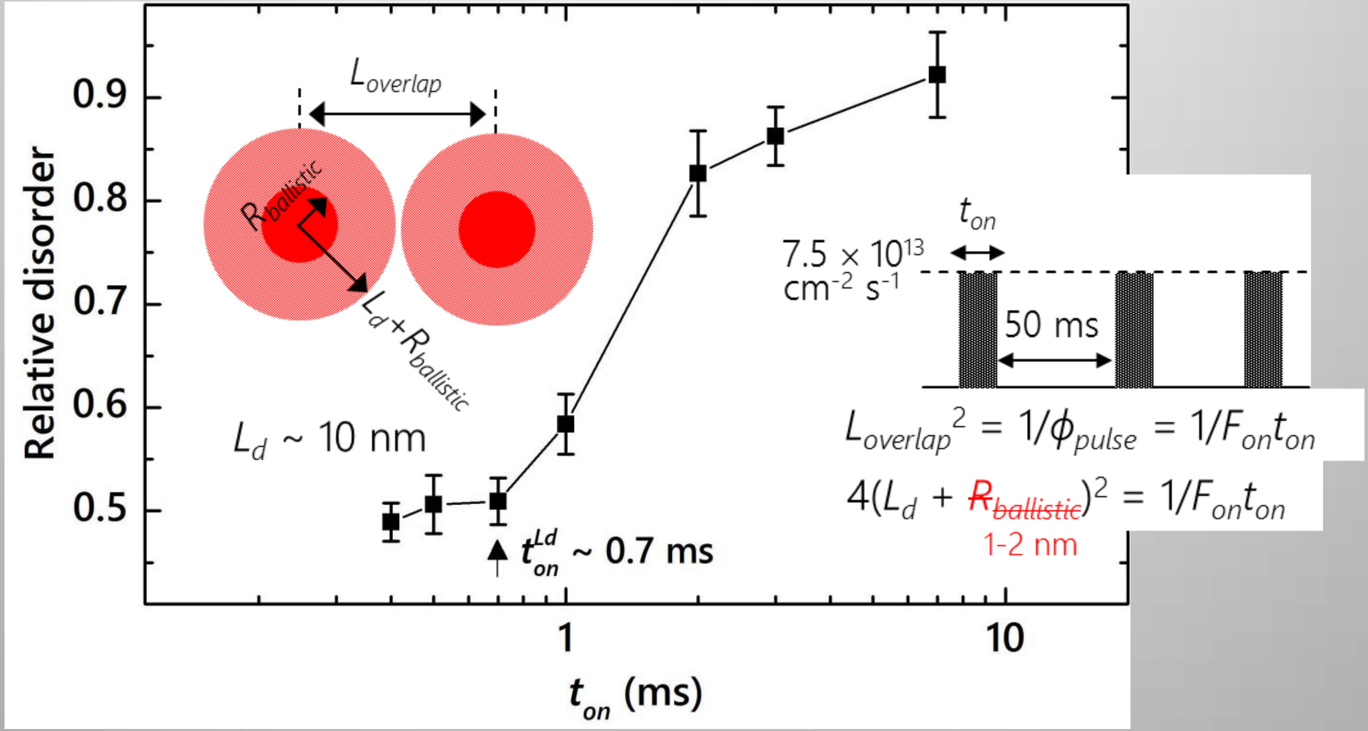
$$L_{\text{overlap}} \approx \frac{1}{\sqrt{F_{\text{on}} t_{\text{on}}}}$$

$$L_{\text{overlap}} < 2L_{\text{d}}$$

JAP 118, 135709 (2015)

For SiC @100 C (500 keV Ar): L_d ~ 10 nm

500 keV Ar ions into 4*H*-SiC



Beam pulsing can be used to access zero-flux irradiation condition!

JPD 49, 19LT01 (2016).

For Si: Monotonic τ(T) dependence

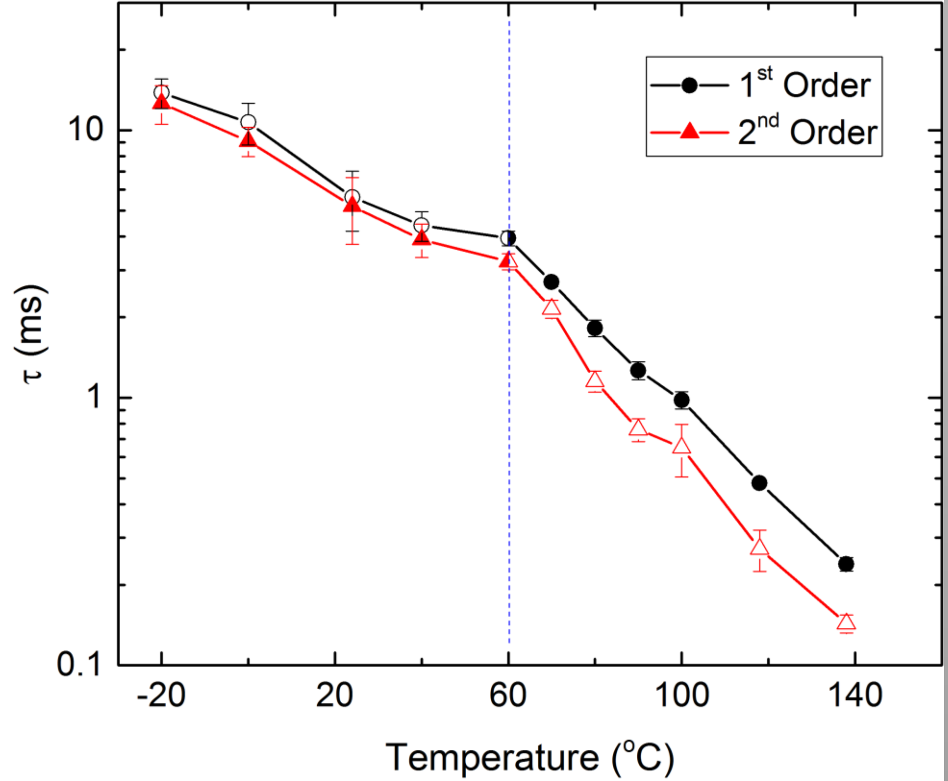
- τ decreases with increasing T

1st order

$$\frac{\partial}{\partial t} n_{\mathrm{d}} \propto n_{\mathrm{d}}$$

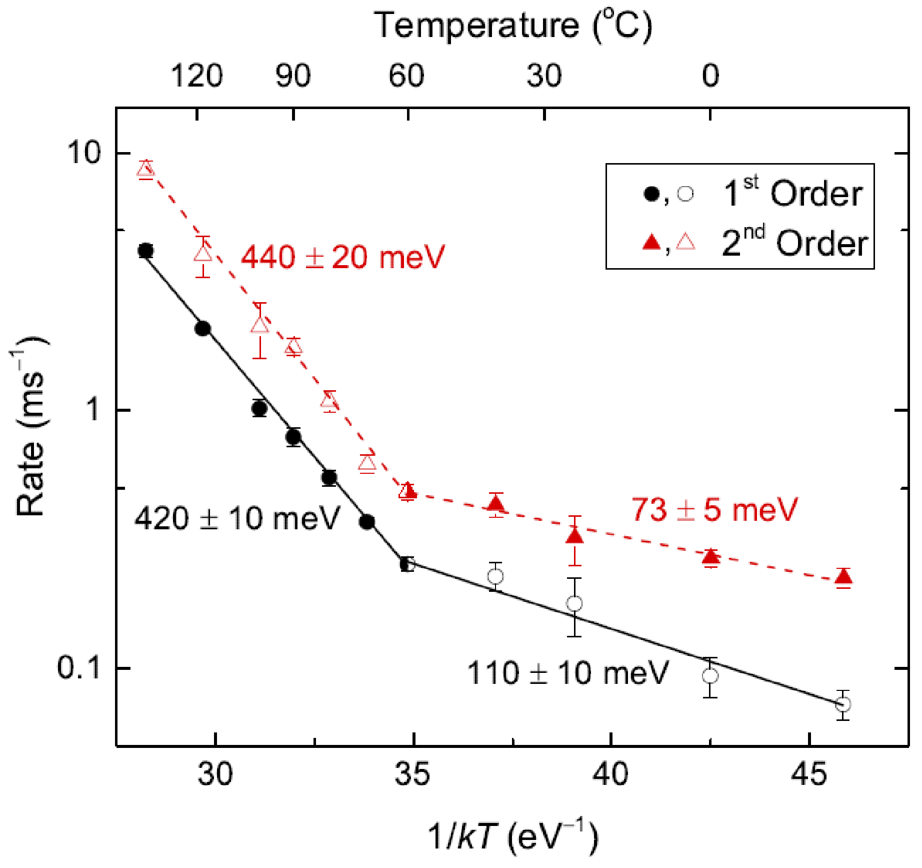
2nd order

$$\frac{\P}{\P t} n_{\mathrm{d}}\ \mu\ n_{\mathrm{d}}^{2}$$



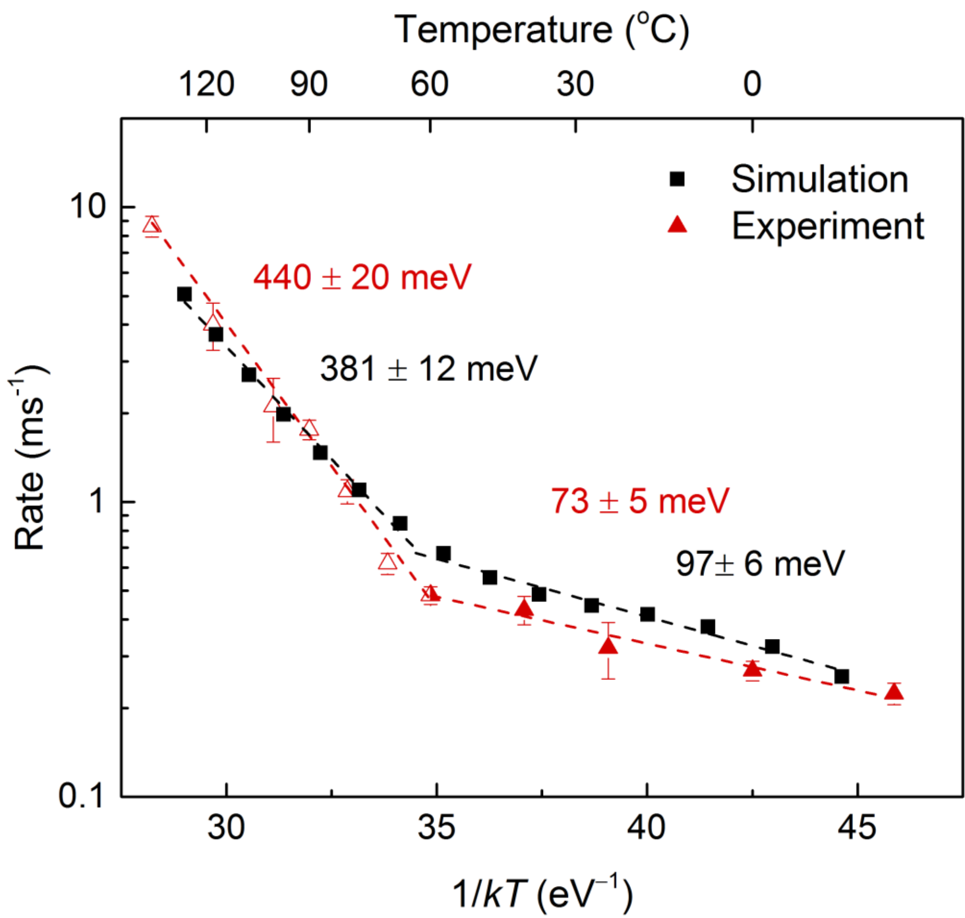
Change in dominant defect interaction mechanism at 60 C

- The existence of two distinct Arrhenius regions with vastly different E_a
- Saturation of the DA efficiency above 60 C
- Change from 2nd order to 1st order fit



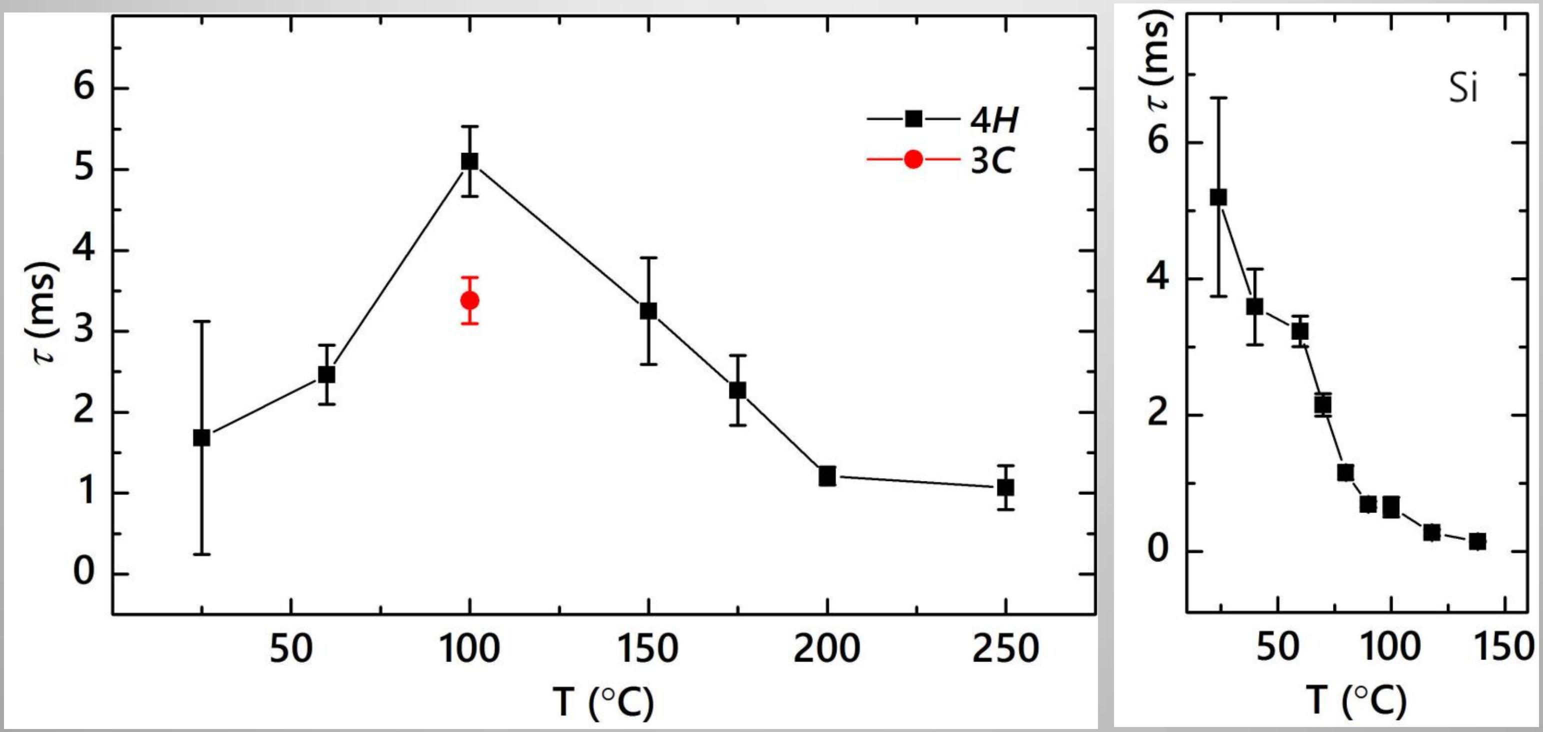
Rate Theory Modeling

- Defect interactions
  - $(I, V) + sink \rightarrow sink$
  - $V + V \rightarrow V_2$
  - $I + V_2 \rightarrow V$
- Diffusion Coefficients
  - $D_{v,i} = \gamma_{v,i}^{m} \lambda^2 e^{-E_{v,i}^{m}/k_b T}$
- Activation energies
  - Vacancy migration: 0.4 eV
  - Interstitial migration: 0.1 eV



A non-monotonic τ(T) dependence for SiC

500 keV Ar ions into 4*H*- and 3*C*-SiC

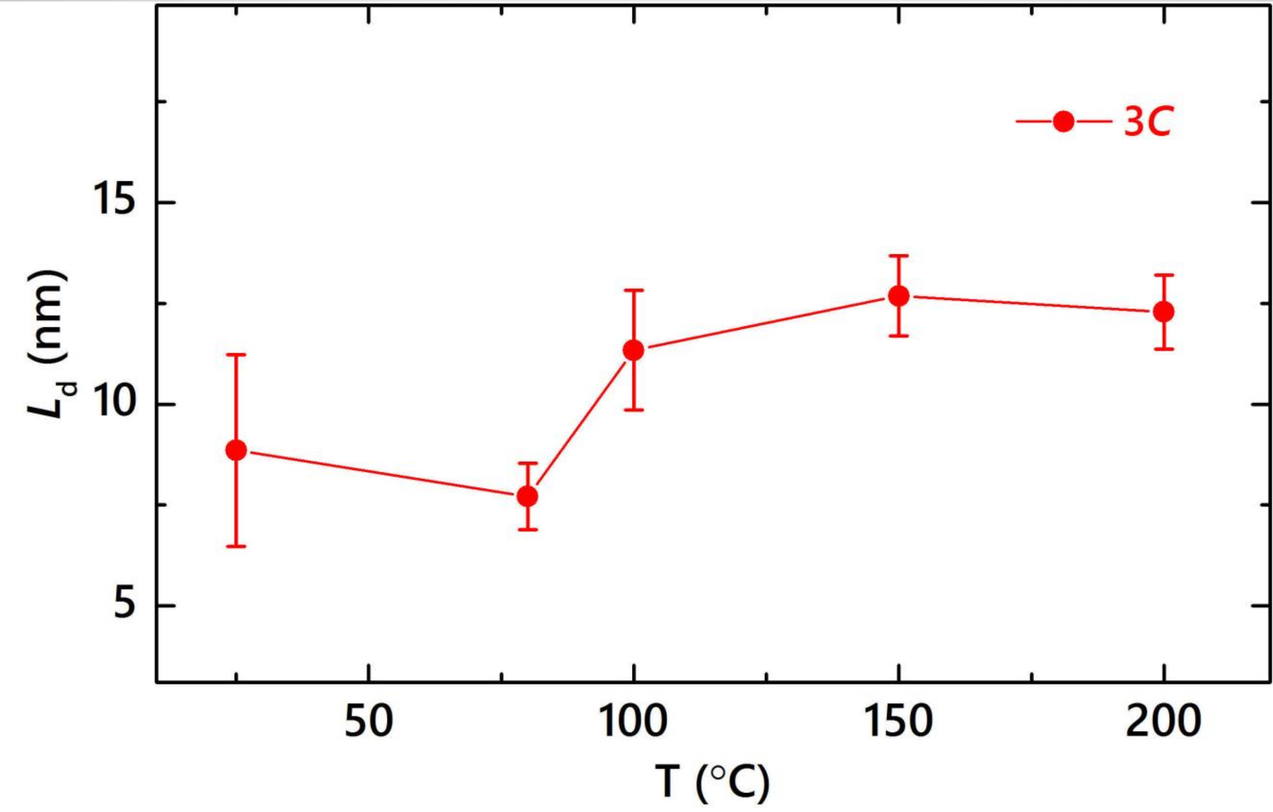


The dynamic annealing mechanism changes at 100 °C

APL 106, 202102 (2015). Sci. Rep. 6, 30931 (2016).

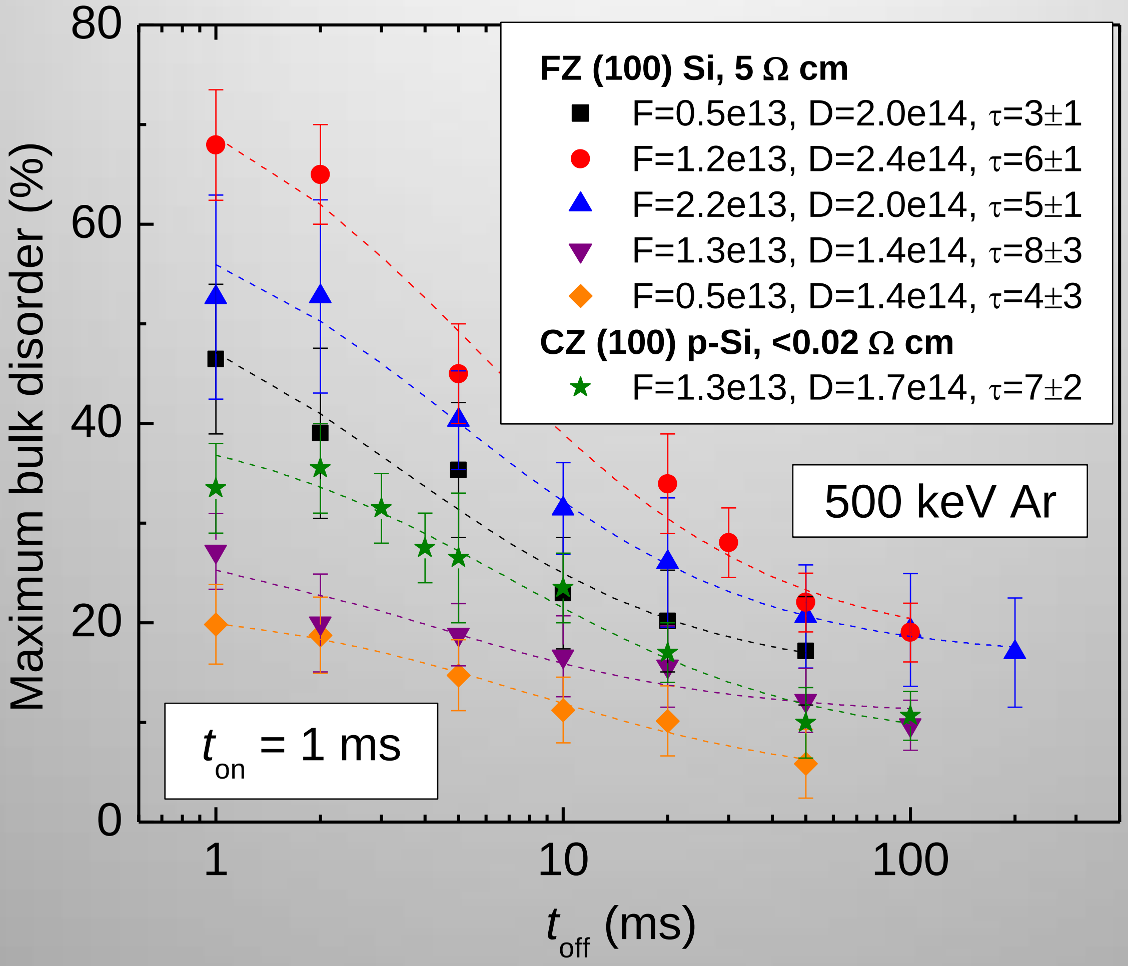
L_d is weakly dependent on T for SiC

500 keV Ar ions into 3C-SiC



JPD 49, 19LT01 (2016).

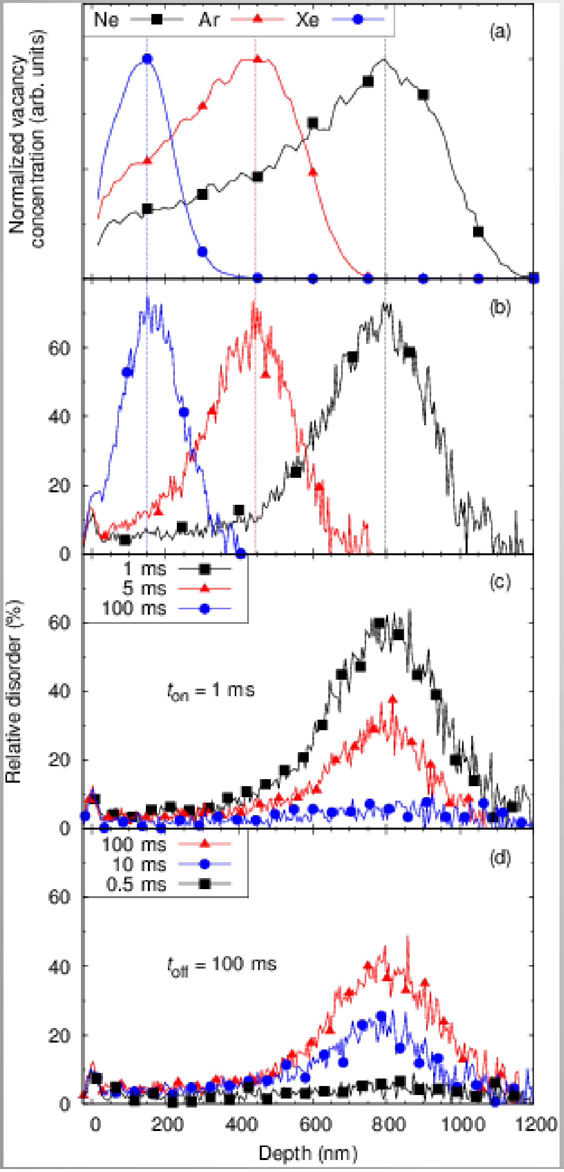
For Si @RT: τ is essentially independent of maximum beam flux, dose, or dopants

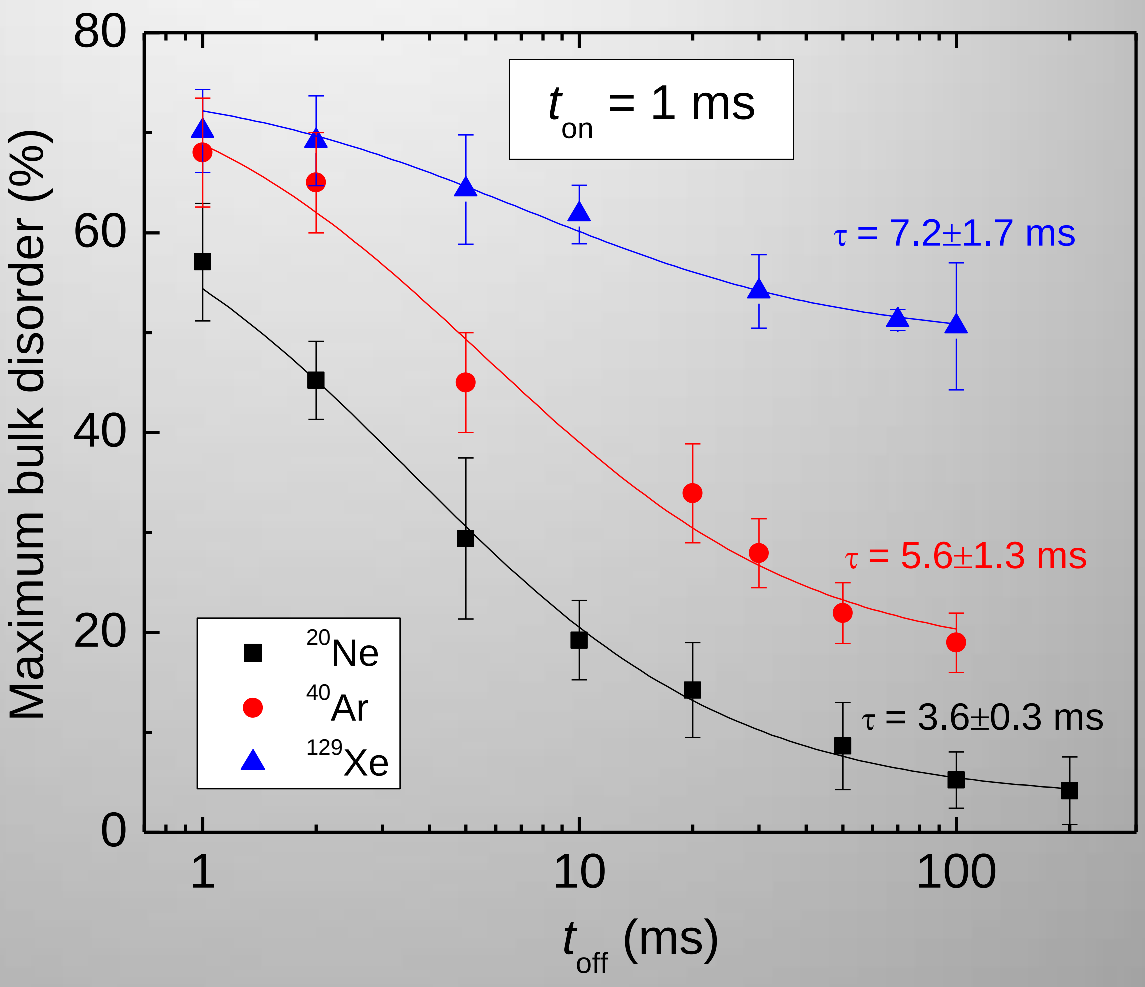


$$n_d = n_{¥} + \frac{n_0 - n_{¥}}{1 + \frac{t_{off}}{t}}$$

JAP 118, 135709 (2015)

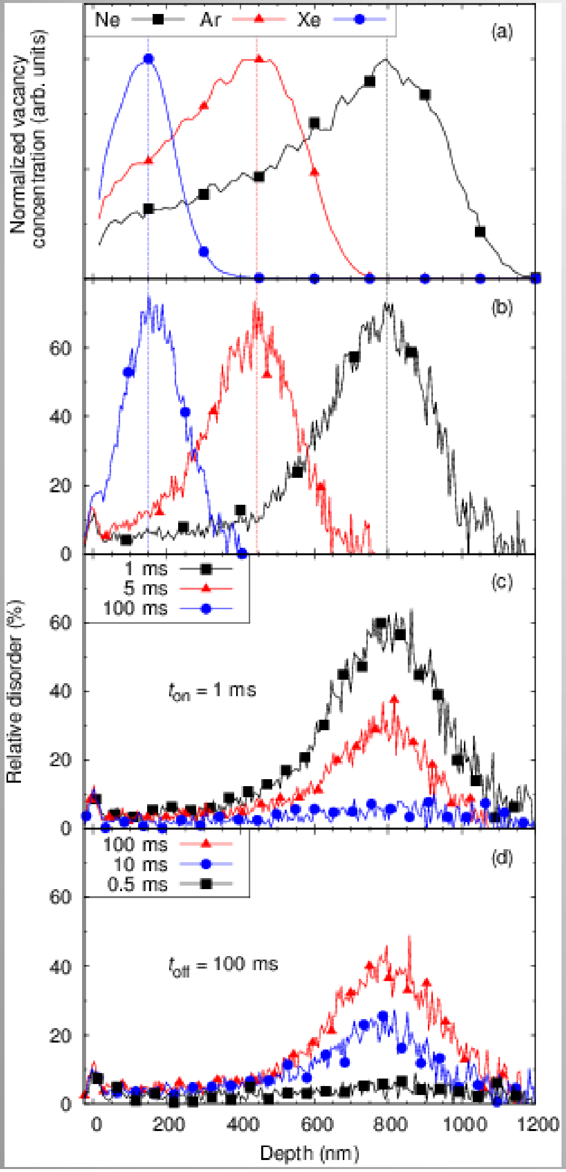
For Si @RT: τ depends on cascade density

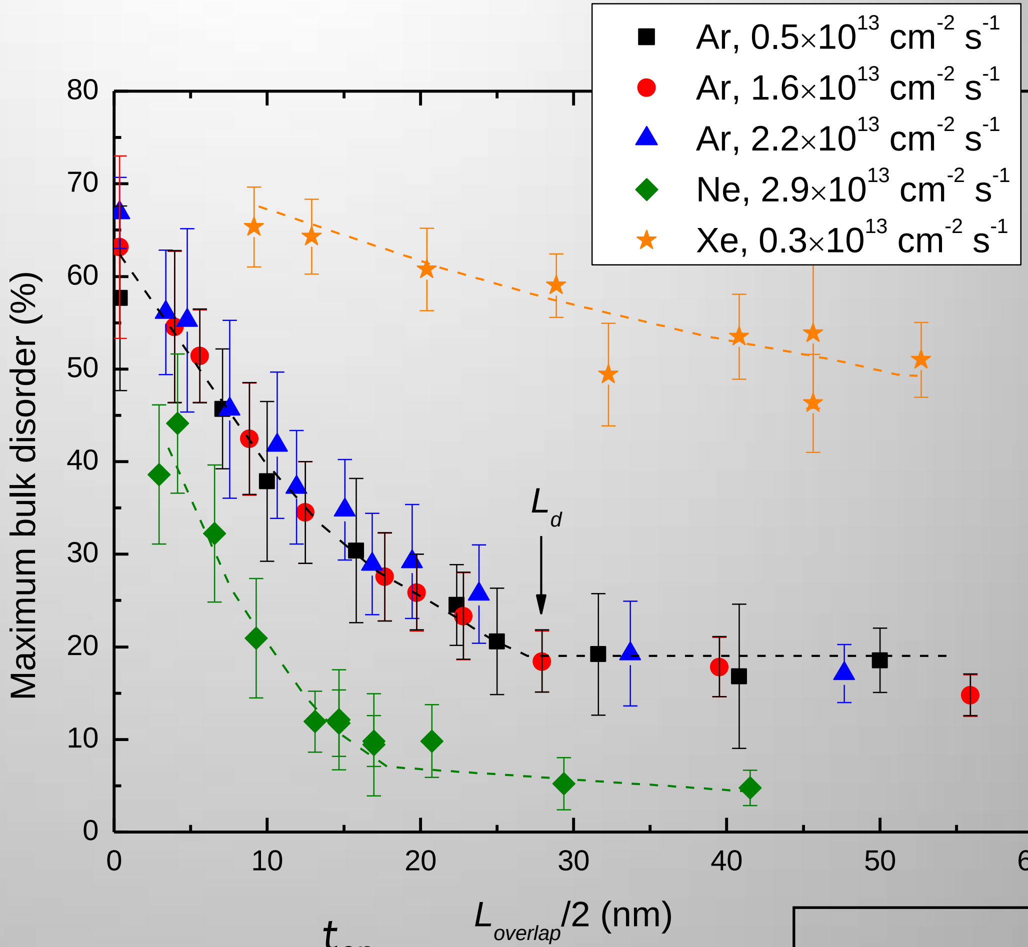


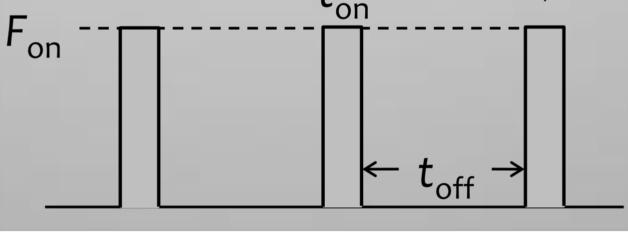


JAP 118, 135709 (2015)

For Si @RT: L_d depends on cascade density





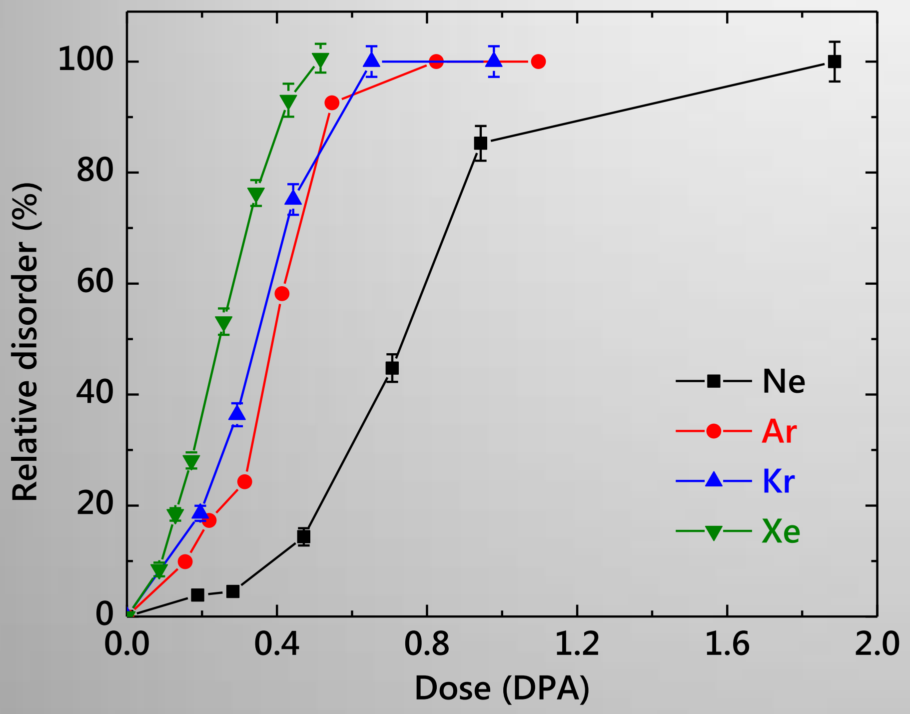


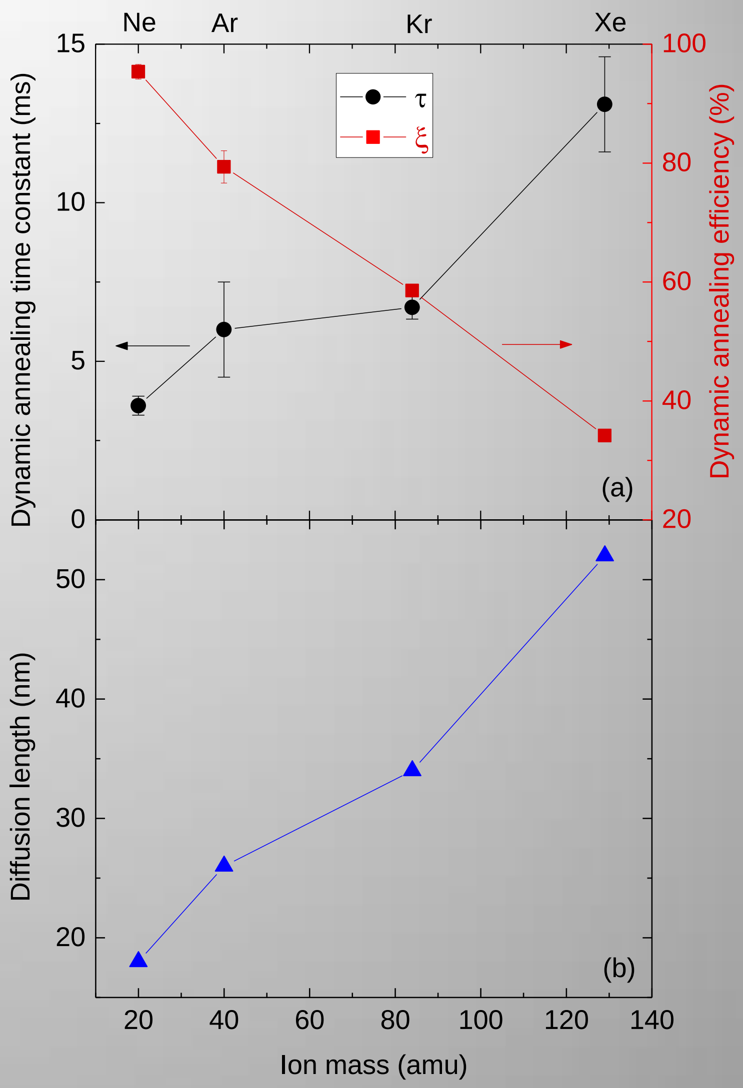
$$L_{overlap} \approx \frac{1}{\sqrt{F_{on} t_{on}}}$$

$$L_{overlap} < 2L_d$$

JAP 118, 135709 (2015)

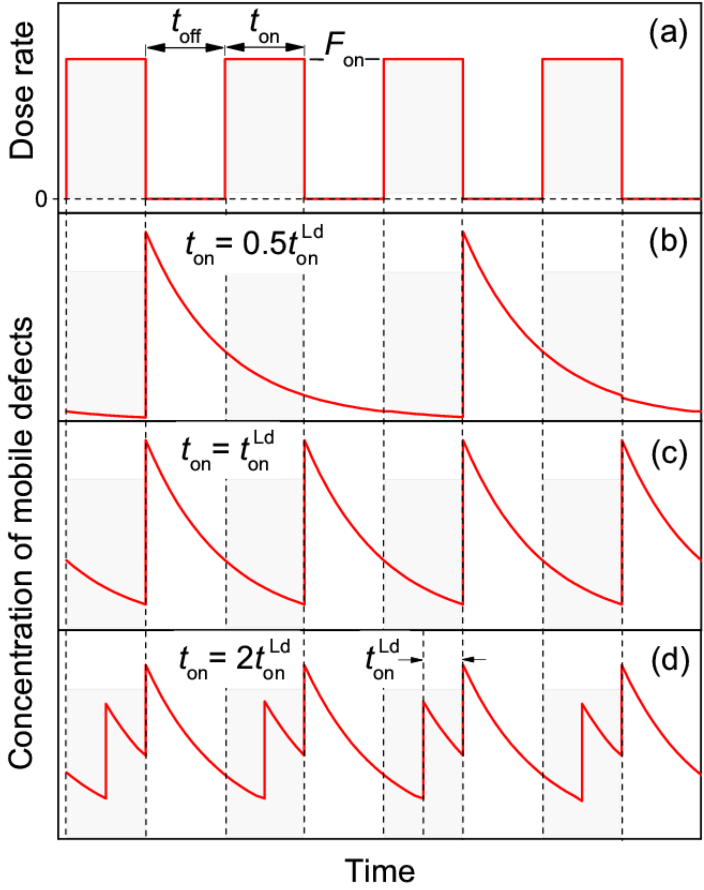
Strong cascade density effects in Si @RT

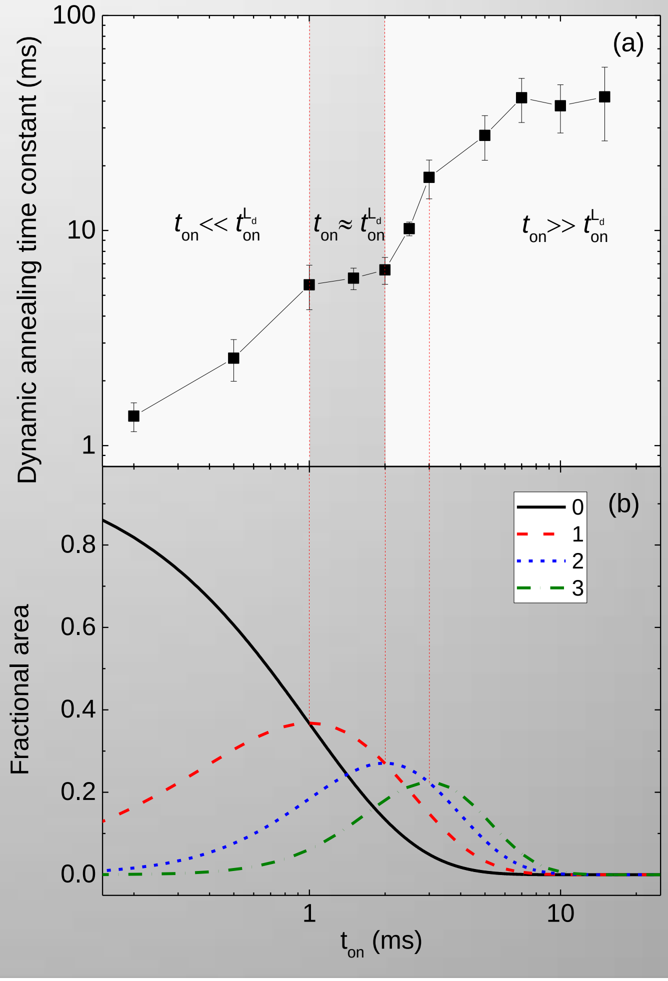




JAP 118, 135709 (2015)

Insight into instantaneous defect concentration effects





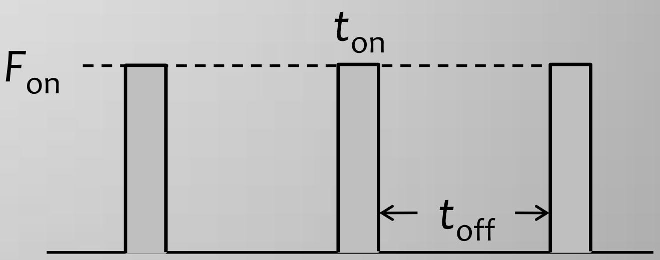
JAP 118, 135709 (2015)

Summary

- We are developing a pulsed beam method to access radiation defect dynamics. Our method
  - Separates spatial (L_d) and temporal (τ) information
  - Does not require assumptions about explicit defect interaction processes for measurements of τ and L_d
  - Averages over multiple cascades and pulses
- For SiC, Si, and Ge around RT:

  τ ~ 0.1-100 ms

  L_d ~ 10-50 nm



- Pulsed beam data is used to benchmark damage buildup models
- This method is not limited to semiconductors or single crystals
- Defect dynamics in other nuclear materials? Role of pre-existing defects and interfaces? Role of steady state defect concentrations?

Thank you!

Sergei Kucheyev

(925) 422-5866

kucheyev@llnl.gov

Lawrence Livermore
National Laboratory