Aspects of Multiscale Modeling: Primary Damage and Rate Theory Models

Presented by:

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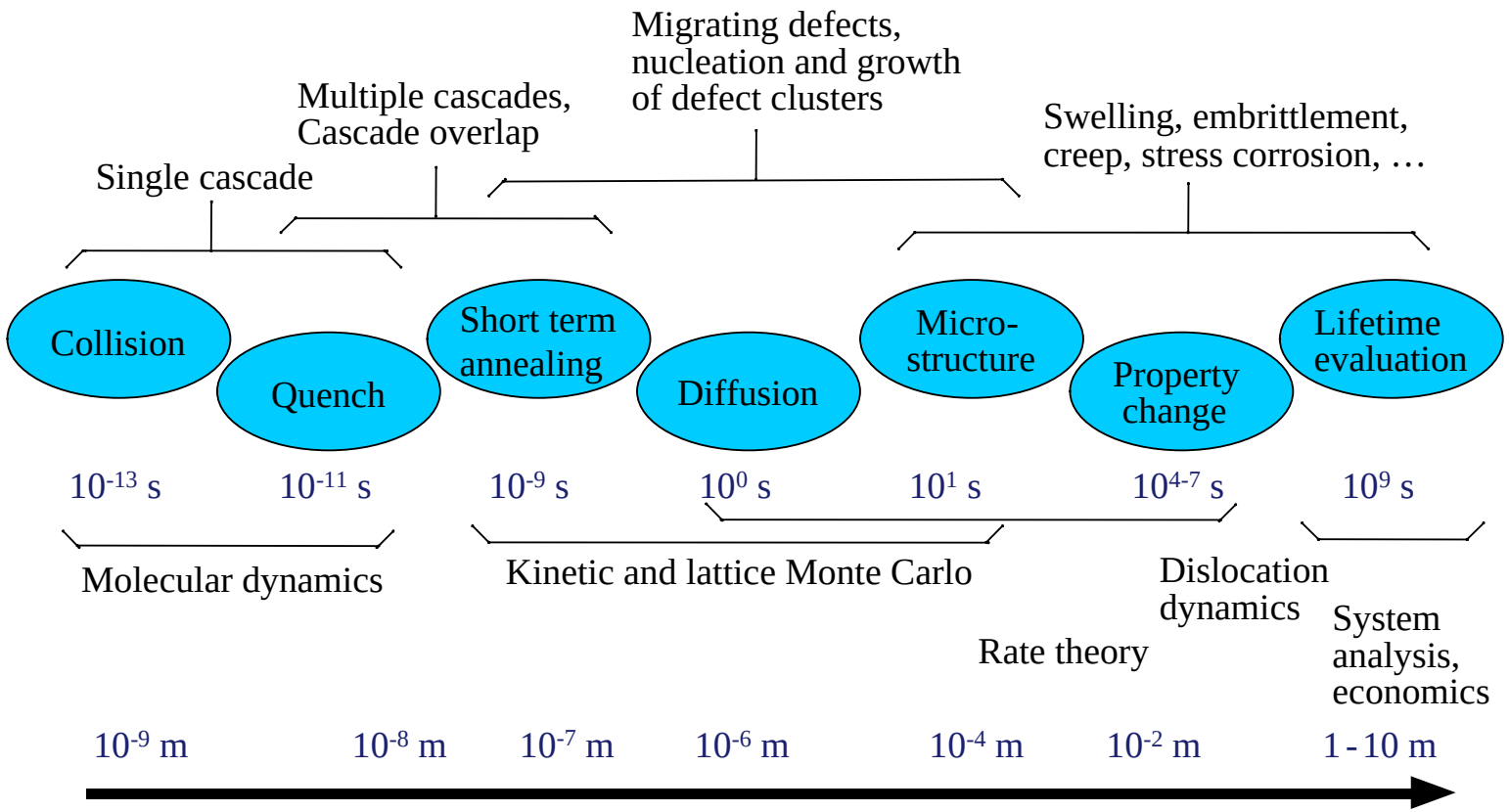
Overview of Presentation

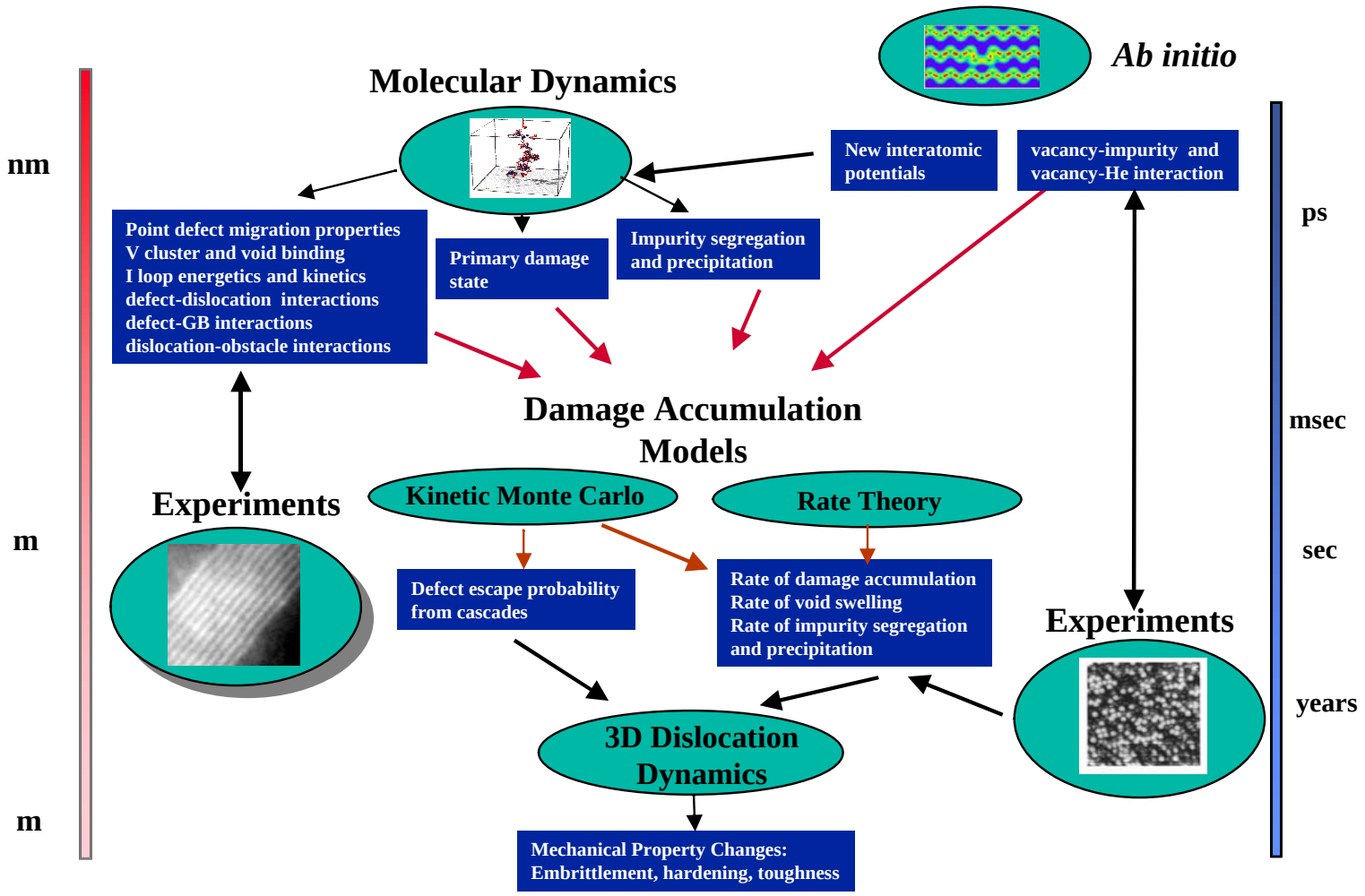
- **Example to illustrate multiscale aspects of radiation damage modeling**
  - -- impact of primary damage parameters in rate theory microstructural evolution model
- **Investigations of primary radiation damage formation: Results and status from molecular dynamics**
  - -- Iron (primarily)
  - -- SiC
- **Investigations of radiation-induced microstructural evolution: Results and status from rate theory**
  - -- status of comprehensive model developed for austenitic stainless steels, evaluation of temperature-change experiment
  - -- helium effects on cavity evolution, austenitic steels and vanadium
- **Modeling focus on radiation effects since this topic is of limited interest to materials research community**
  - -- results of relevant research from broader community monitored and incorporated
  - -- work has been supported in part by other programs, primarily the DOE Office of Basic Energy Sciences and the Nuclear Regulatory Commission
  - -- common research interest between these programs and need to leverage limited funding
  - -- aspects relevant to DT fusion damage production emphasized

Radiation damage simulations require understanding of physical phenomena across many length and time scales



A range of methods are required to examine the relevant phenomena

Multiscale modeling approach



Application of MD-derived parameters in kinetic models used to predict microstructural evolution

Cascade survival efficiency, η, is a key parameter in rate equations used to calculate vacancy (C_v) and interstitial (C_i) concentrations:

$$\frac{dC_{i,v}}{dt} = \eta G_{dpa} - \alpha C_i C_v - D_{i,v} C_{i,v} S^T_{i,v}$$

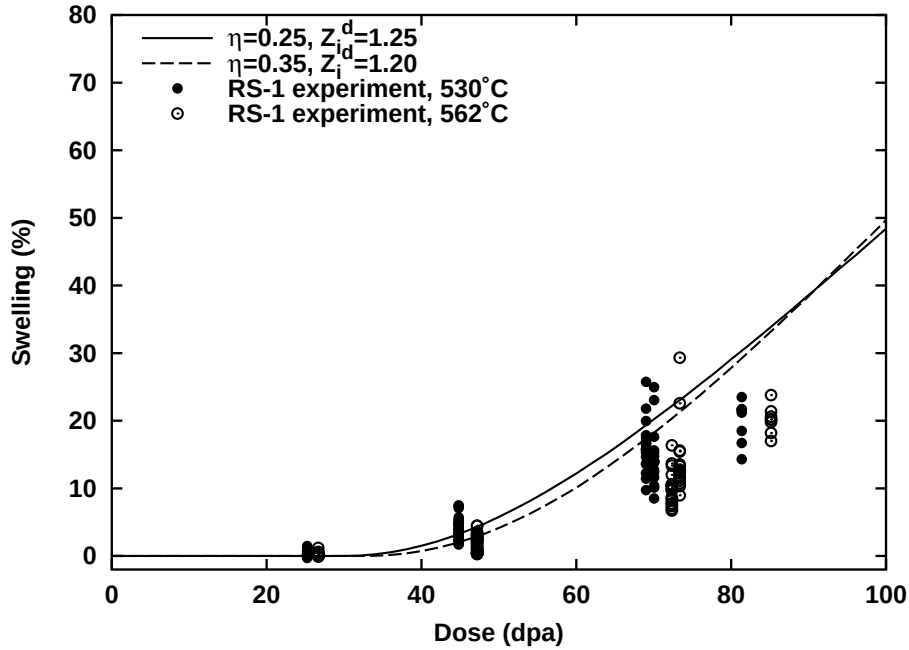
For a simple example of point defect absorption dominated by dislocations with sink strength S_d, swelling rate is proportional to product of η and net dislocation bias (Z_i^d-1):

$$\frac{dV}{dt} = \frac{\eta G_{dpa}}{S_v^d}\left(Z_i^d - 1\right)$$

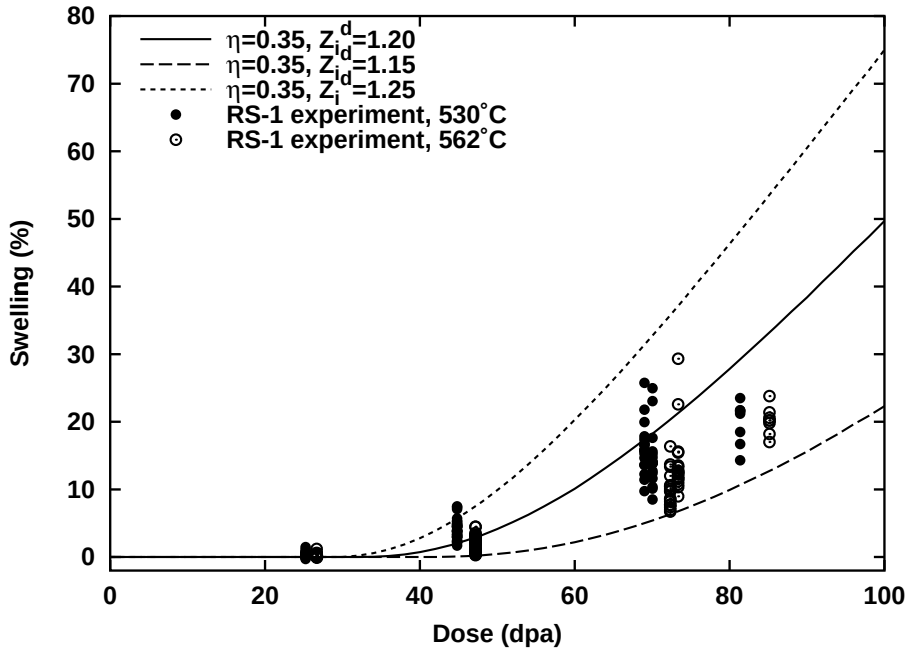
In this case, data fitting can not be used to obtain unique set of model parameters. MD cascade simulations provide independent estimate of η and thereby permit better estimate of Z_i^d.

Comparison of model predictions and swelling data from FFTF irradiation of 20% CW 316 stainless steel

Effect of varying η and Z_i^d

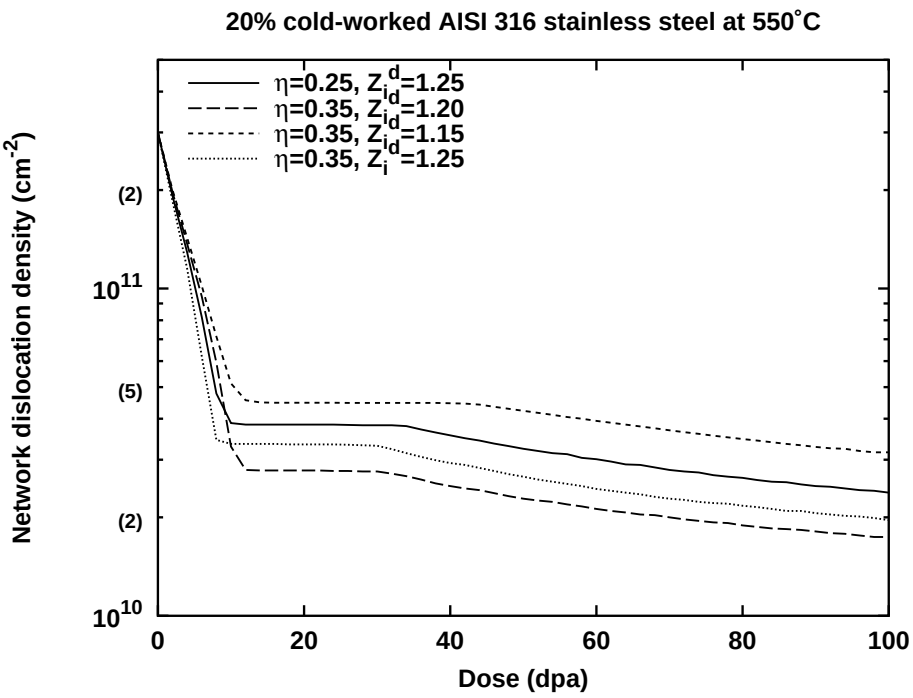


MD calculated spectrum-average value η=0.35



Co-evolution of all microstructural components important for predicting any one component in complex model

- **Influence of MD parameter variations on predicted network dislocation density in AISI 316 stainless steel 550˚C**



Investigations of primary radiation damage formation: Results and status from molecular dynamics

- **most extensive investigation of primary damage has been in iron**
  - -- MD simulations carried out using the MOLDY code by Finnis (AEA Report, 1988)
  - -- Interatomic potential used is EAM-type by Finnis and Sinclair (Phil. Mag. 1984)
  - -- modified for cascade simulations by Calder and Bacon (JNM 1993)
  - -- constant pressure, periodic boundary condition
  - -- boundary atoms not damped, results in some heating
  - -- no electronic losses or electron-phonon coupling, hence energy of cascade simulation analogous to damage energy in NRT displacement model:

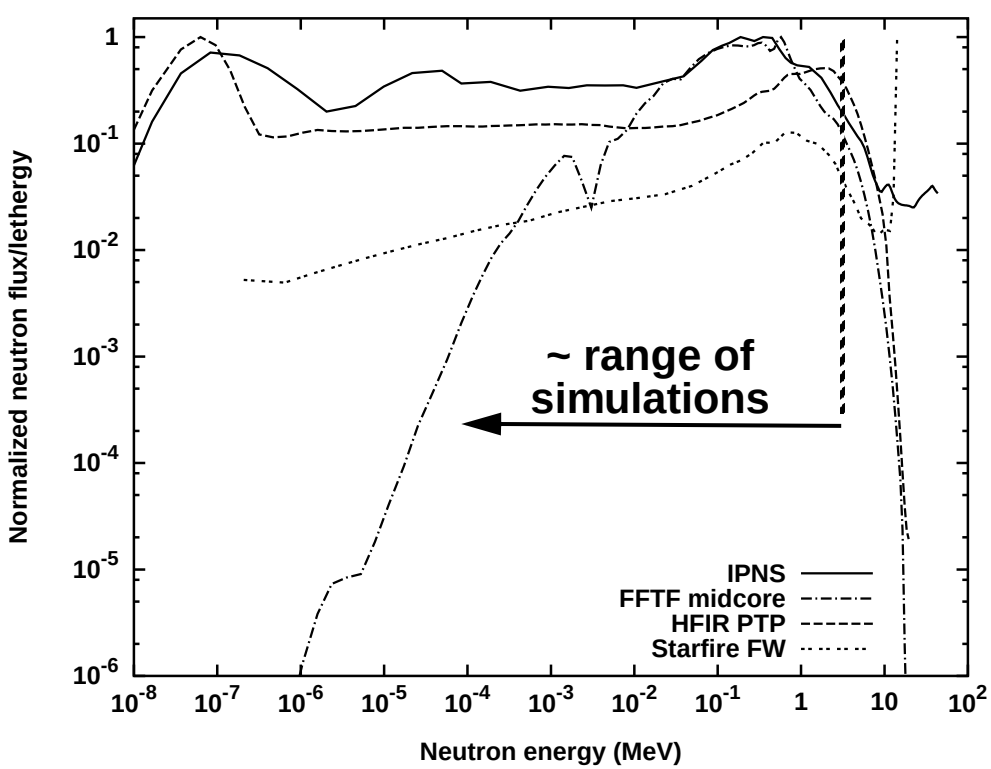
$$E_{MD} \sim T_d \text{ (NRT)} < E_{PKA}$$

MD cascade data base covers neutron energies relevant to fission reactors: demonstrated relevance to fusion neutron spectrum

$E_{neutron}$ (MeV)	Average E_{PKA} (keV)	E_{MD} (keV)	n_{NRT} (displacements)	atoms in simulation
0.0034	0.116	0.1	1	3456
0.036	1.24	1	10	54k
0.074	2.54	2	20	54k
0.19	6.61	5	50	128k
0.40	13.7	10	100	250k
0.83	28.8	20	200	250k
1.8	61.3	40	400	1024k
2.3	78.7	50	500	2.249M
5.09	175.8	100	1000	5.031M

Typical neutron energy spectra

ORNL DWG 97-1258(



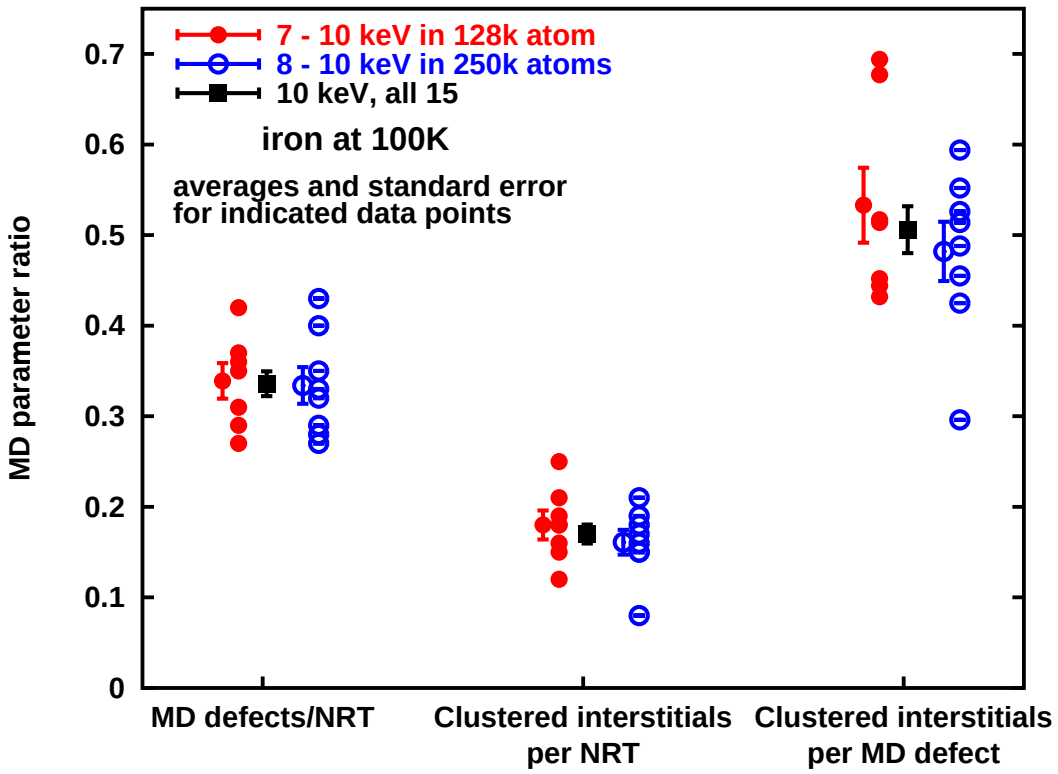
Summary of current 100K MD cascade database

E_{MD} (keV)	number of simulations completed	90% confidence factor[1]
0.1	40	1.645
0.2	32	“
0.3	128	“
0.5	20	1.731
1.	8	1.895
2	8	“
5.	8	“
10.	15	1.762
20.	10	1.834
30.	10	“
40.	8	1.895
50.	9	1.860
100	8	1.895

[1] 90% confidence interval on mean values is obtained by multiplying standard error of mean by this value

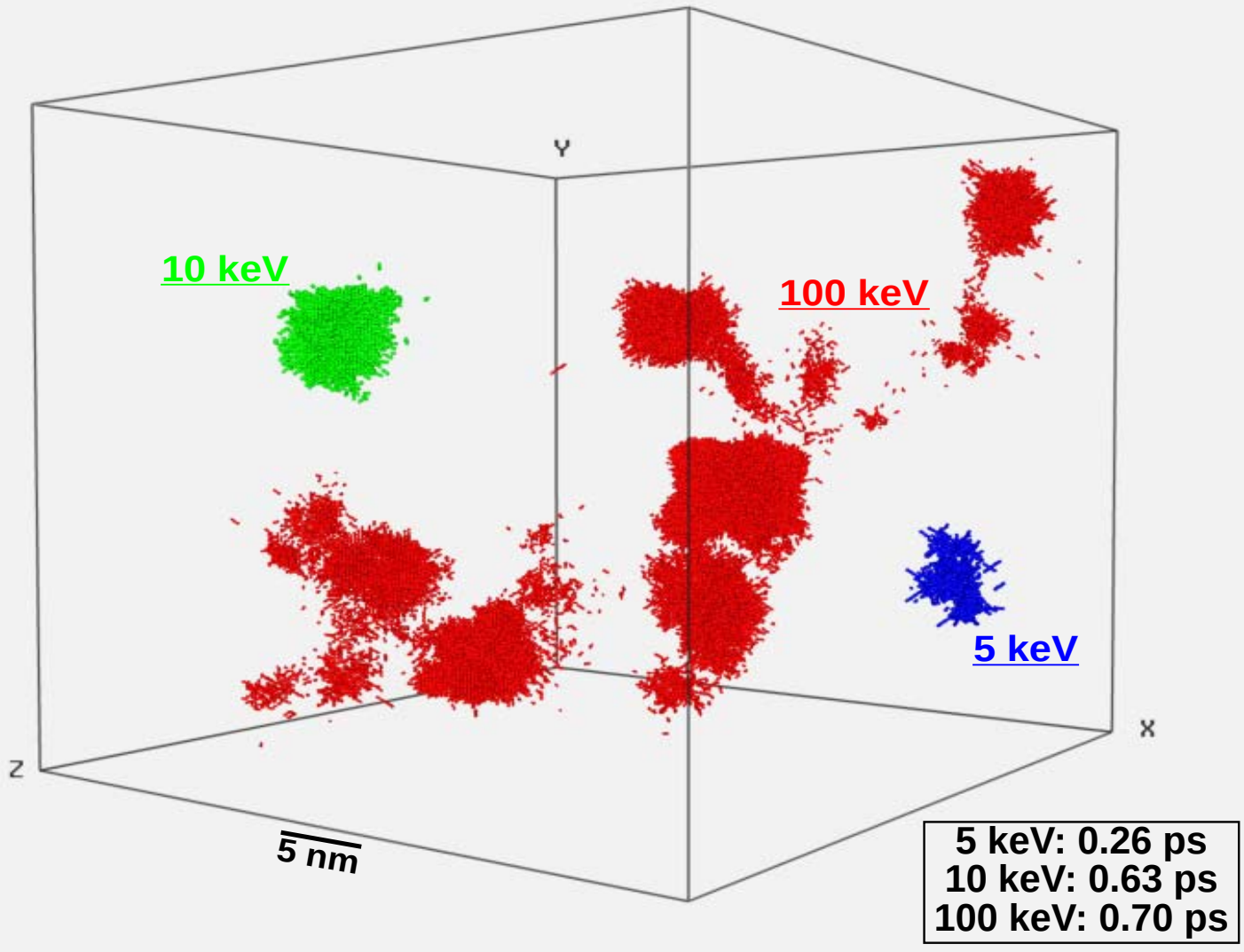
<insert 20 keV cascade movie>

Statistical variation in 10 keV cascade simulations



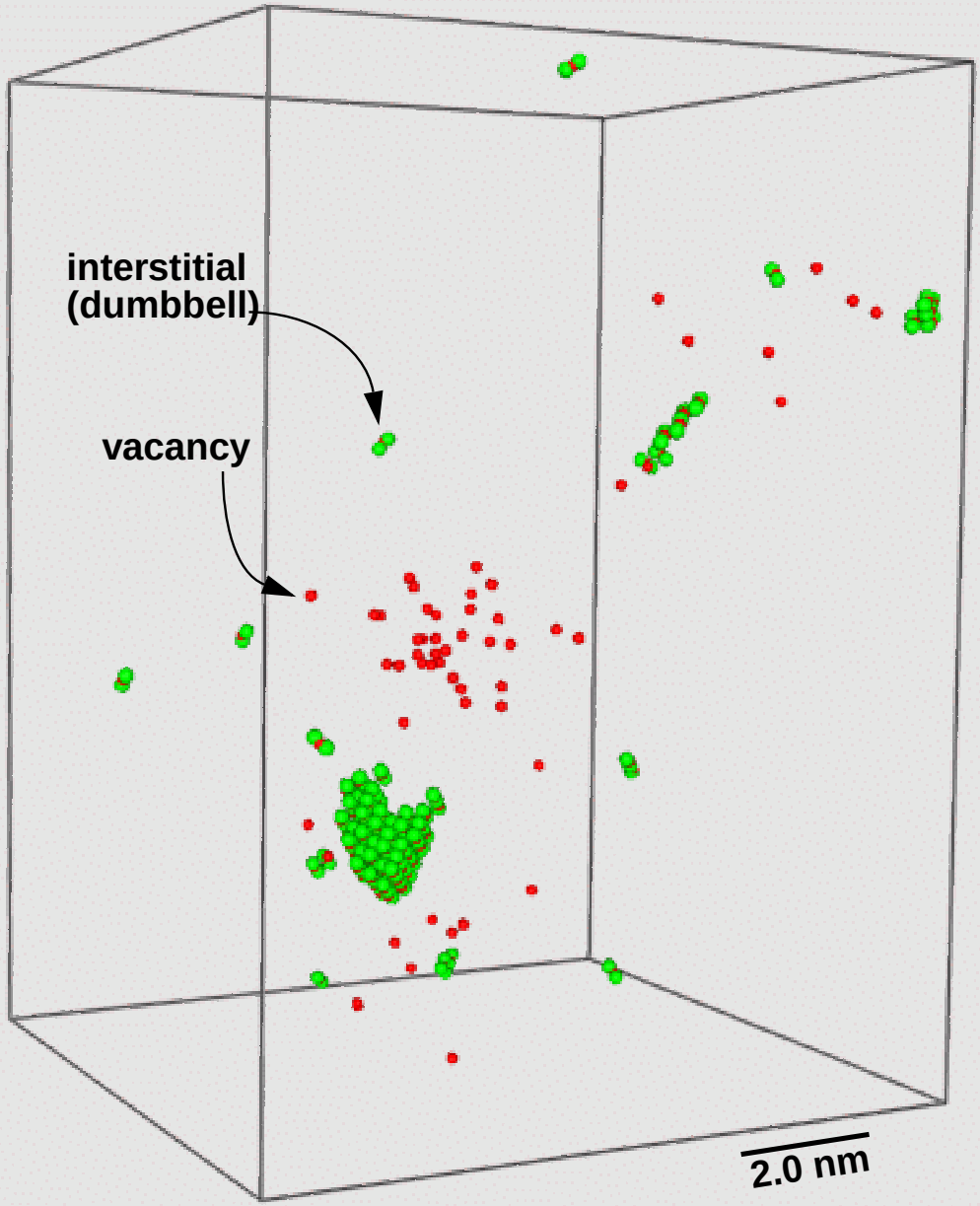
- valid averages are obtained with fewer (~7) simulations at the higher energies
- more effective averaging arises from events that involve many more atoms, truly cascade-like
- little effect of simulation cell size observed when box is “large enough”
- slight difference in clustering averages may be due to heating, residual T~250K for 250k atoms and ~350K for 128k atoms

MD Cascade Simulations in Iron at 100K



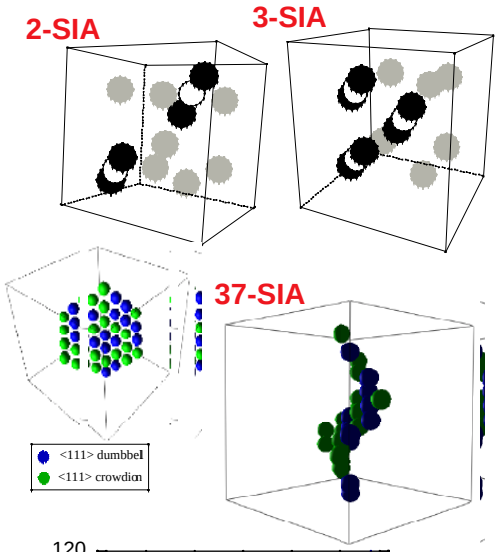
Residual Defects from 20 keV Cascade in Iron at 600K

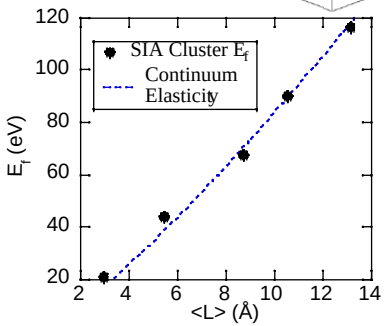
55 surviving Frenkel pair, 28% of NRT
84% of interstitials in clusters (one with 33 interstitials)



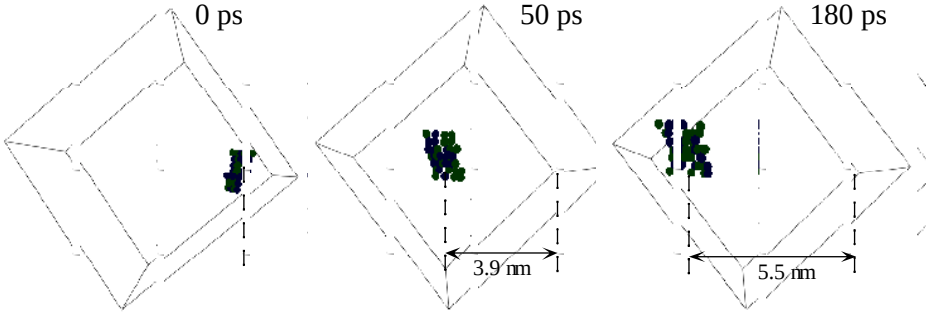
Self-interstitial clusters: <111> loops

- **Stability of <111> self-interstitial atom (SIA) clusters revealed by recent atomistic modeling (*Finnis-Sinclair and EAM-type interatomic potentials*)**



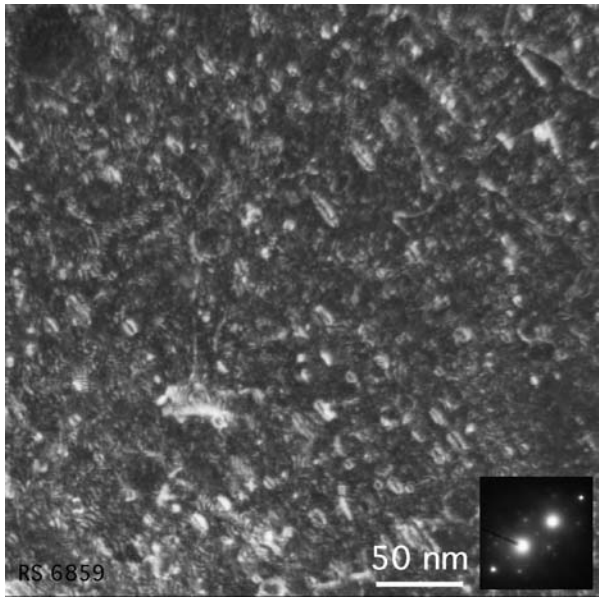


MD simulations at T=560 K reveal rapid (E^m < 0.1 eV) 1-D migration



- Form highly kinked, proto-<111> dislocation loops directly in cascades
- Migrate in 1-dimension with high mobility

Self-interstitial clusters: <100> loops



Irradiated martensite steel (Fe-8Cr)

(experimental micrograph by R. Schäublin)

- **Large number density of <100> loops observed (TEM)**
- **Previous model (Eyre and Bullough) suggests formation from faulted Frank loops (<110> platelets):**

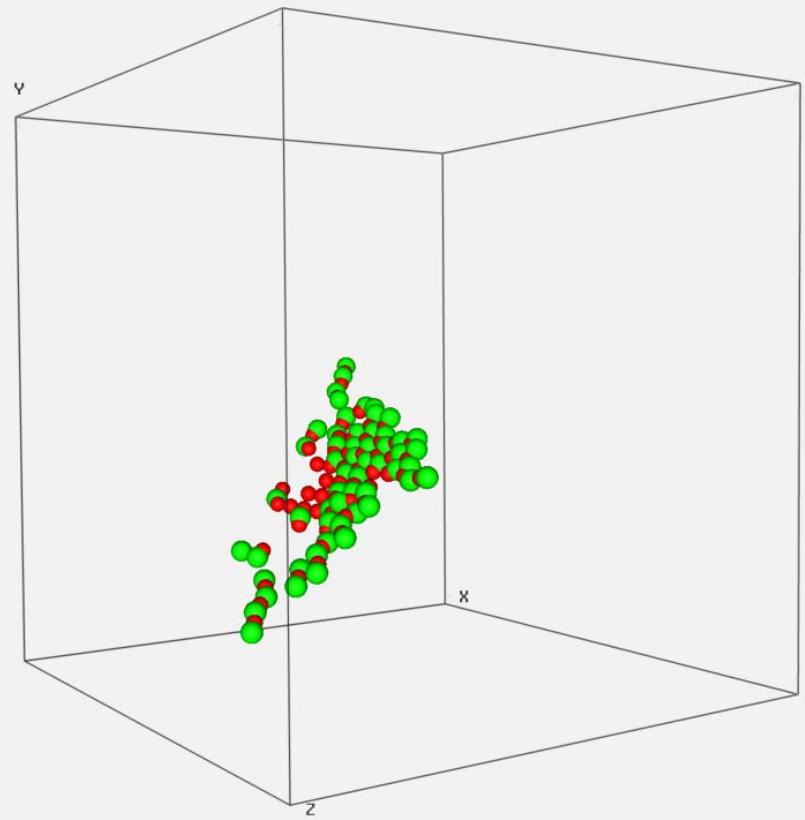
$$\frac{a_o}{2}[110] + \frac{a_o}{2}[\bar{1}10] \rightarrow a_o[010]$$

- New insight: <100> loops form through interaction of highly mobile, <111> loops:

$$\frac{a_o}{2}[111] + \frac{a_o}{2}[1\bar{1}\bar{1}] \rightarrow a_o[100]$$

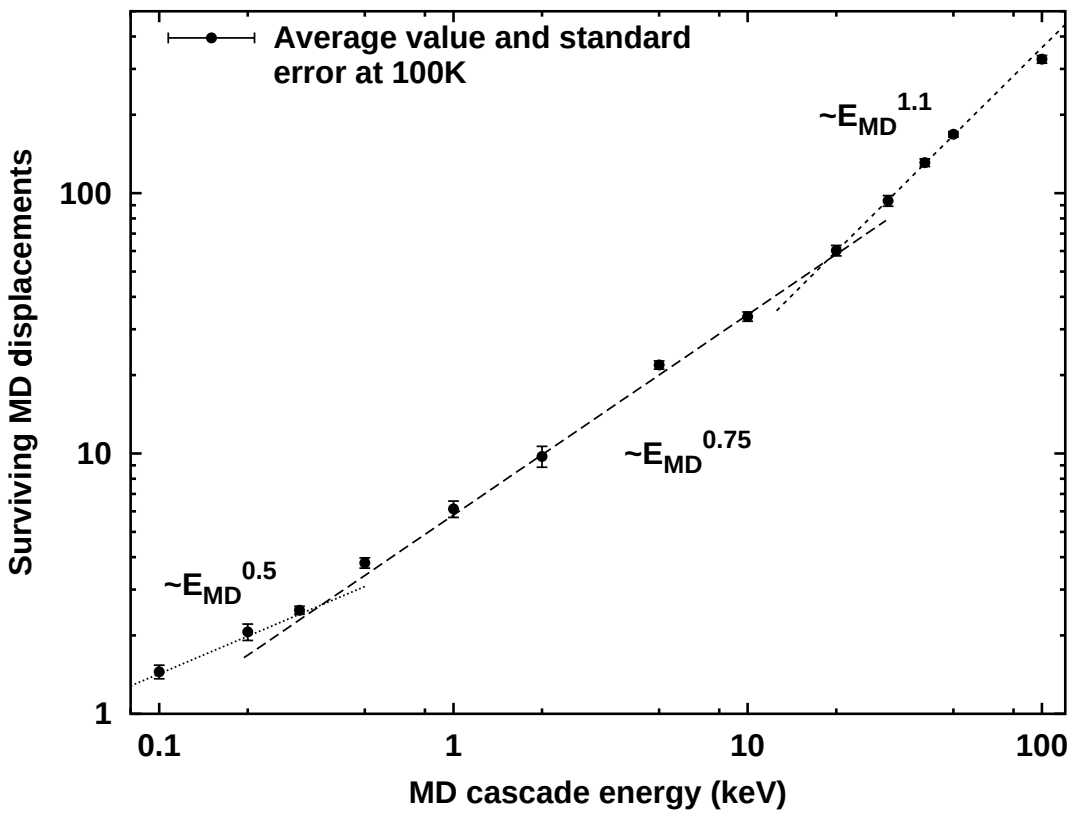
Mechanism for enhanced defect production discovered at low energies

300 eV PKA at 100K **[114] PKA direction**



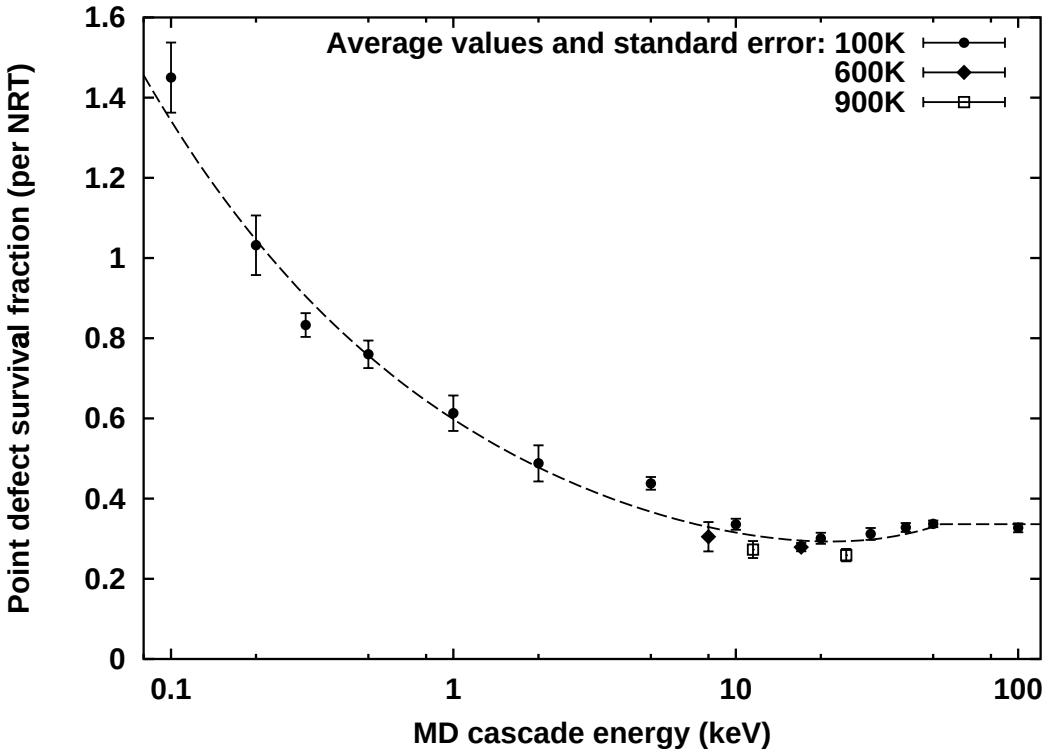
- **if PKA direction lies in {110} plane, the tendency for atomic defects to be confined to a single, close-packed plane can be referred to as "planar channeling"**
- **leads to higher than average stable point defect production as the interstitials and vacancies are created with greater separation**
- **for 300 eV cascades, survival increased by 60% for [114] cascades relative to high index direction**

Cascade energy dependence of MD primary damage parameters: Total number of surviving displacements



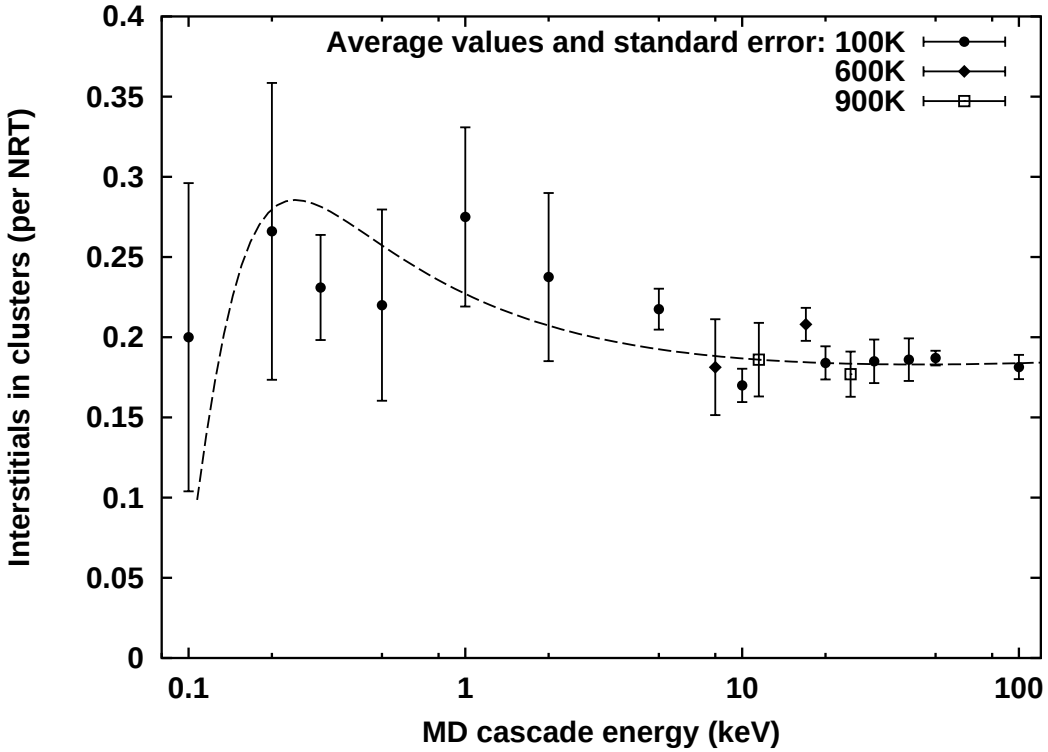
- **stable defect formation exhibits three regions with different energy dependence**
  - -- low: efficient production of isolated defects
  - -- intermediate: compact cascades
  - -- high: well isolated subcascades, enhanced defect survival between subcascades
- **NRT model, defect production** $\propto T_{dam}^{1.0}$

Cascade energy dependence of MD primary damage parameters: Surviving MD displacements per NRT displacement



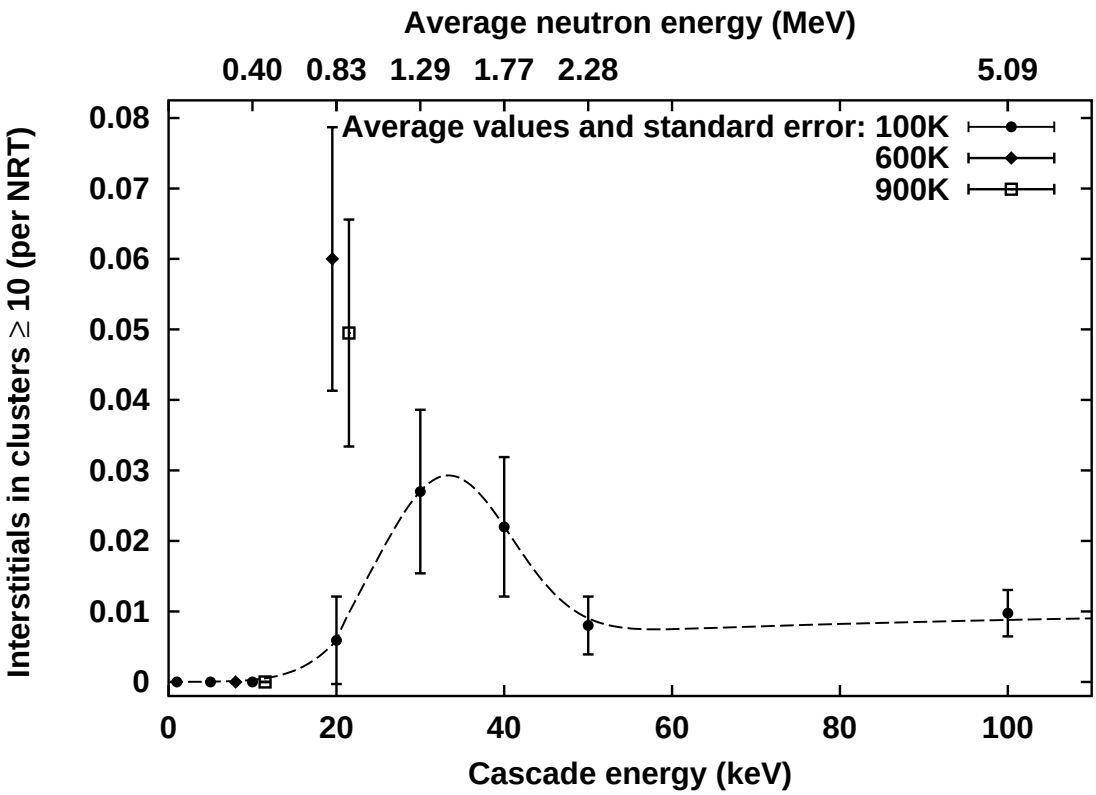
- well behaved, simple analytical fit obtained
- minimum at ~20 keV appears statistically significant
- smooth extrapolation to 100 keV indicates that subcascade effect has saturated

Cascade energy dependence of MD primary damage parameters: clustered interstitials per NRT displacement



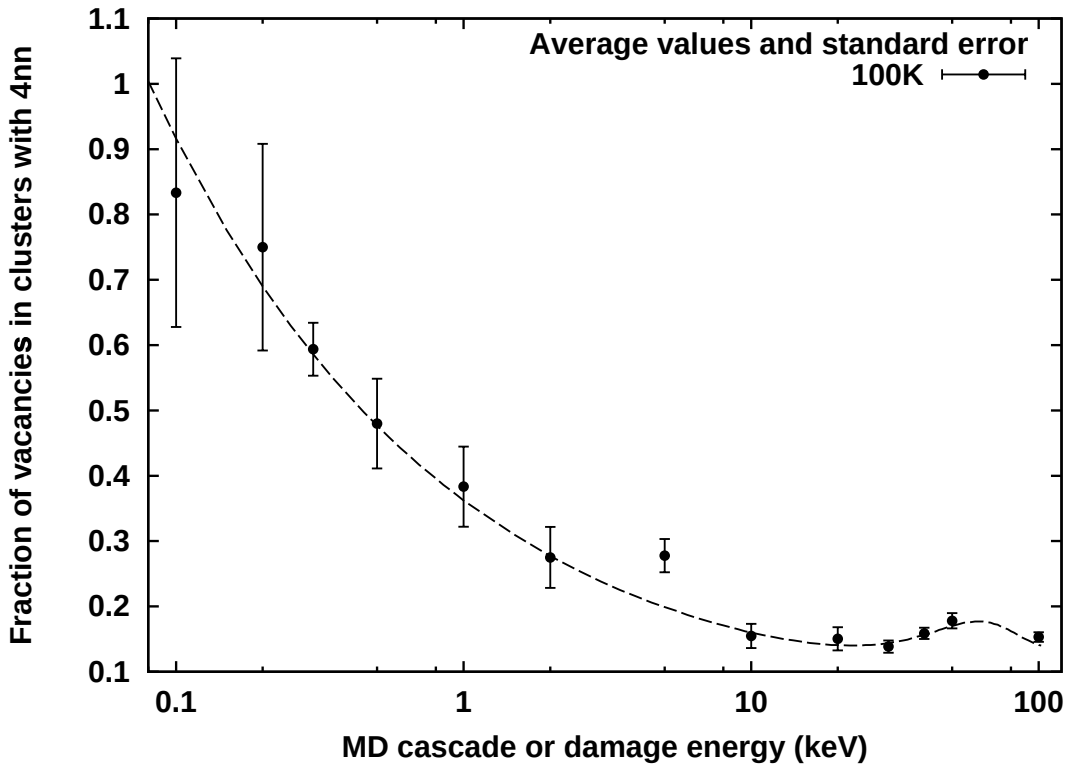
- data scatter leads to larger standard errors
- very weak energy dependence

Cascade energy dependence of MD primary damage parameters: interstitials in relatively large in-cascade clusters (per NRT displacement)



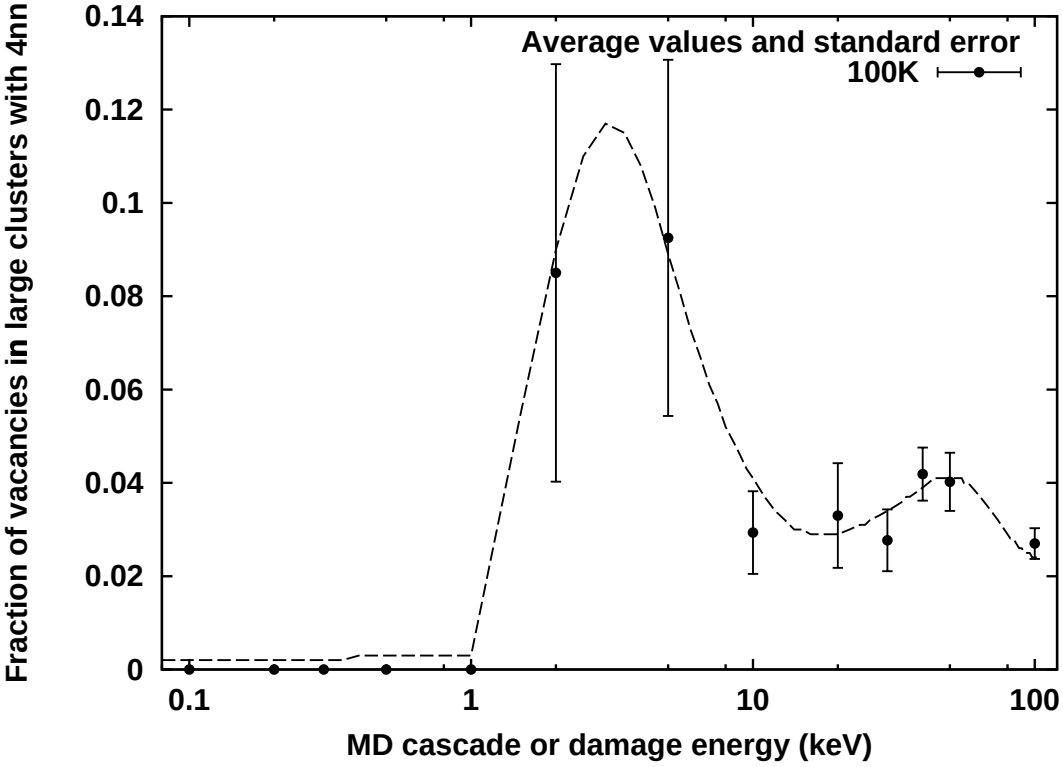
- although standard errors are large, the apparent peak could be due to a maximum average energy density associated with subcascade breakup
- could contribute to a weak spectral effect

Cascade energy dependence of MD primary damage parameters: total clustered vacancies per NRT displacement based on 4nn criterion



- energy dependence of vacancy clustering stronger than that of interstitials, similar shape to total defect survival
- for 4nn clustering criterion, high-energy clustering fraction is somewhat less than that of interstitials

Cascade energy dependence of MD primary damage parameters: fraction of vacancies in clusters per NRT displacement based on 4nn criterion

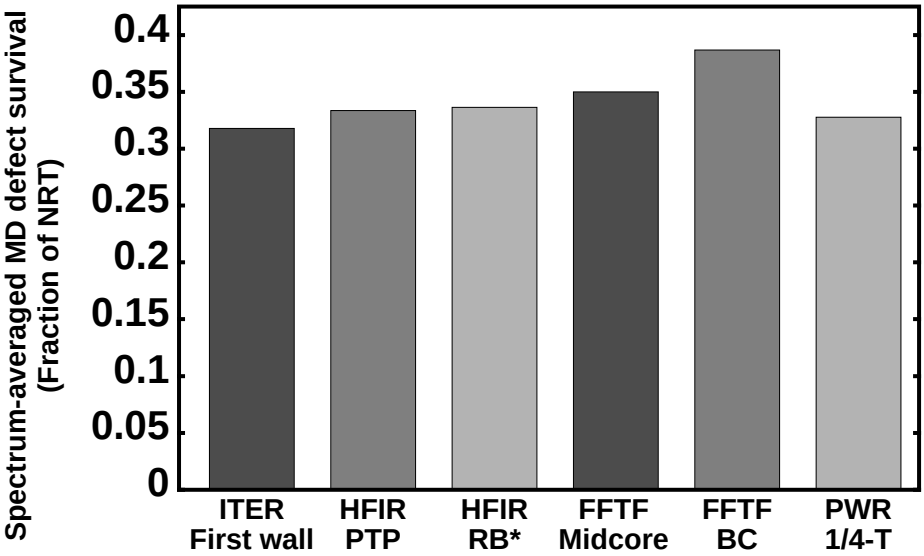


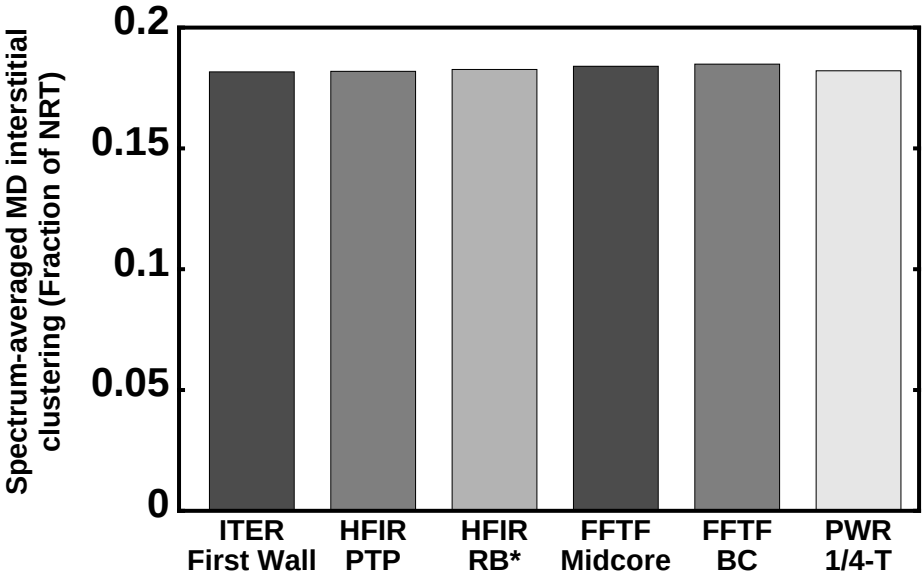
- 5-vacancy cluster was used as criterion for “large” cluster evaluation since vacancy clusters as large as 10 are rarely observed
- energy threshold at lower energy than for interstitial clusters greater than 10
- energy threshold at ~1 keV could contribute to spectrum effect
- even for 4nn criterion, threshold remains lower than for interstitials

Application of MD Results to Neutron Energy Spectrum Effects

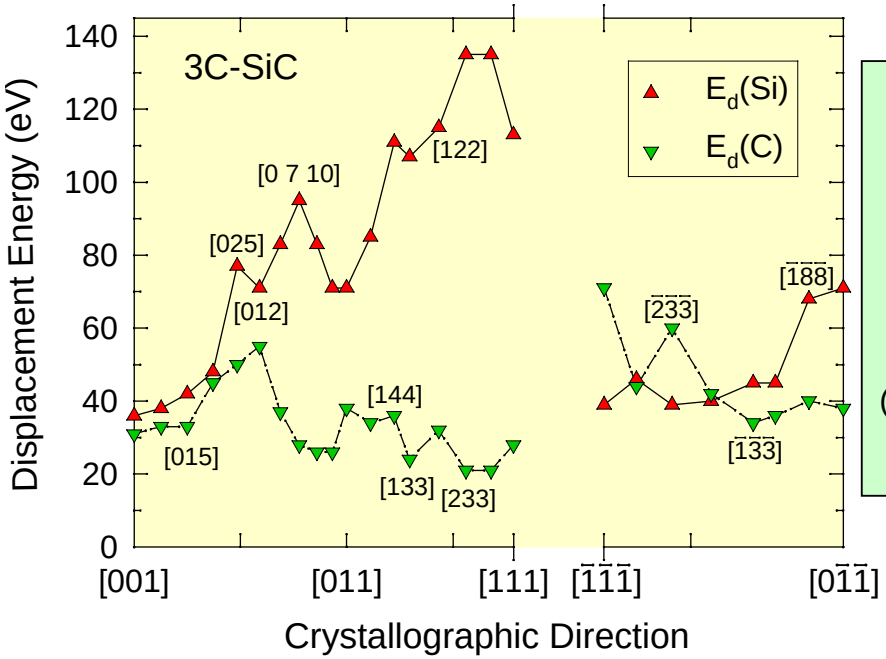
- energy dependence of surviving defect fraction and interstitial clustering fractions was expressed using simple functions obtained by least-squares fitting of results up to 50 keV
- these functions were used to obtain effective damage cross sections and neutron energy-spectrum-averaged defect production cross sections for various reactor environments (SPECOMP/SPECTER code system)
- neutron-energy-spectrum averaged values of both η and f_{icl} only weakly dependent on neutron spectrum, other damage functions exhibit somewhat more spectrum-dependence
- impact of subcascade formation suggests that neutron energy spectrum effects are probably weak in fission reactor irradiations and that fission reactor data should be relevant to displacement component of fusion environment

Spectrum-averaged, MD-based defect production cross sections





MD Simulations of Displacement Energies in 3C-SiC



Highly anisotropic

Easier to displace C

Similar results for 6H-SiC

Recommendation:

E_d(Si) =35 eV

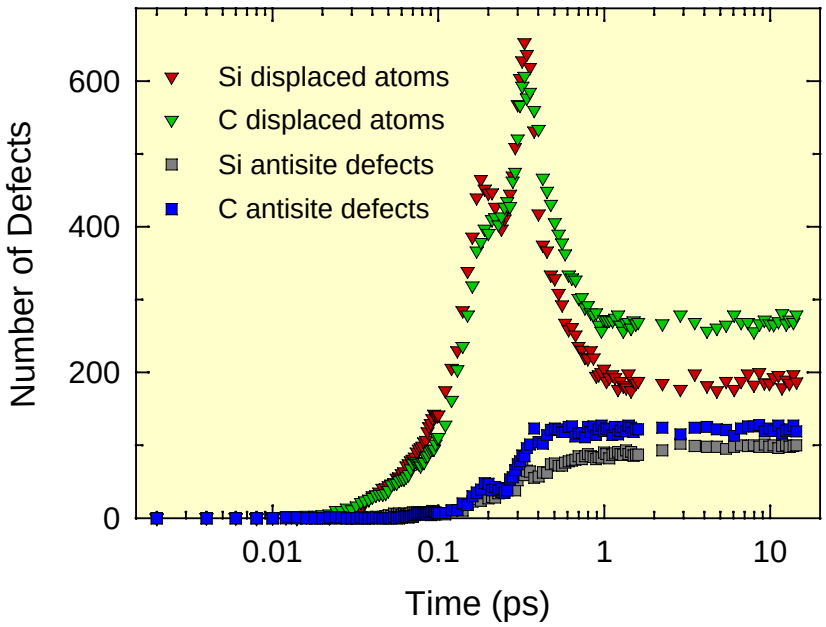
E_d(C)=20 eV

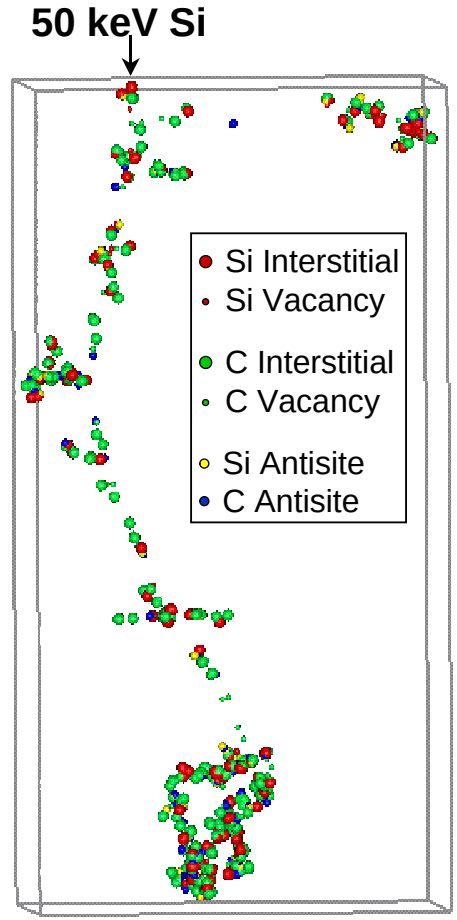
(Consistent with experiments and energetic MD collision cascades)

R Devanathan & WJ Weber, Pacific Northwest National Laboratory
Sponsored by Office of Basic Energy Sciences
Journal of Nuclear Materials 278 (2000) 258-265
Journal of Applied Physics 90 (2001) 2303-2309

Angular dependence of displacement threshold similar to metals, apparent recombination barriers affect measured thresholds at low temperatures

MD Simulation of 50 keV Si Cascade in 3C-SiC





Time dependence and spatial distribution of defect production in a 50 keV Si cascade

F Gao & WJ Weber Pacific Northwest National Laboratory
Sponsored by Office of Basic Energy Sciences
Physical Review B 63 (2000) 054101

Cascade morphology somewhat different, less dense than in metals

Summary of Primary Damage Studies

- **MD cascade simulation energies have reached energies relevant fast neutron irradiation conditions, maximum equivalent neutron energy ~5.1 MeV**
- **Simulations have been completed in sufficient numbers to statistically evaluate the effects of cascade energy and irradiation temperature, database has been reanalyzed using a single set of visualization tools**
- **Above ~10 keV, subcascade formation dominates the details of defect formation**
- **Impact of subcascade formation suggests that neutron energy spectrum effects are probably weak when comparing fission reactor irradiations with DT fusion**
- **Small clusters <111> crowdions (nascent interstitial loops) are highly glissile, E_m~0.1 eV**
- **More work is underway to:**
  - -- verify predictions of EAM-type potentials with ab initio calculations and development of more detailed potentials
  - -- evaluate impact of MD-based primary damage parameters in kinetic models

Brian Wirth:
cascade aging, KMC, SFT-dislocation interactions

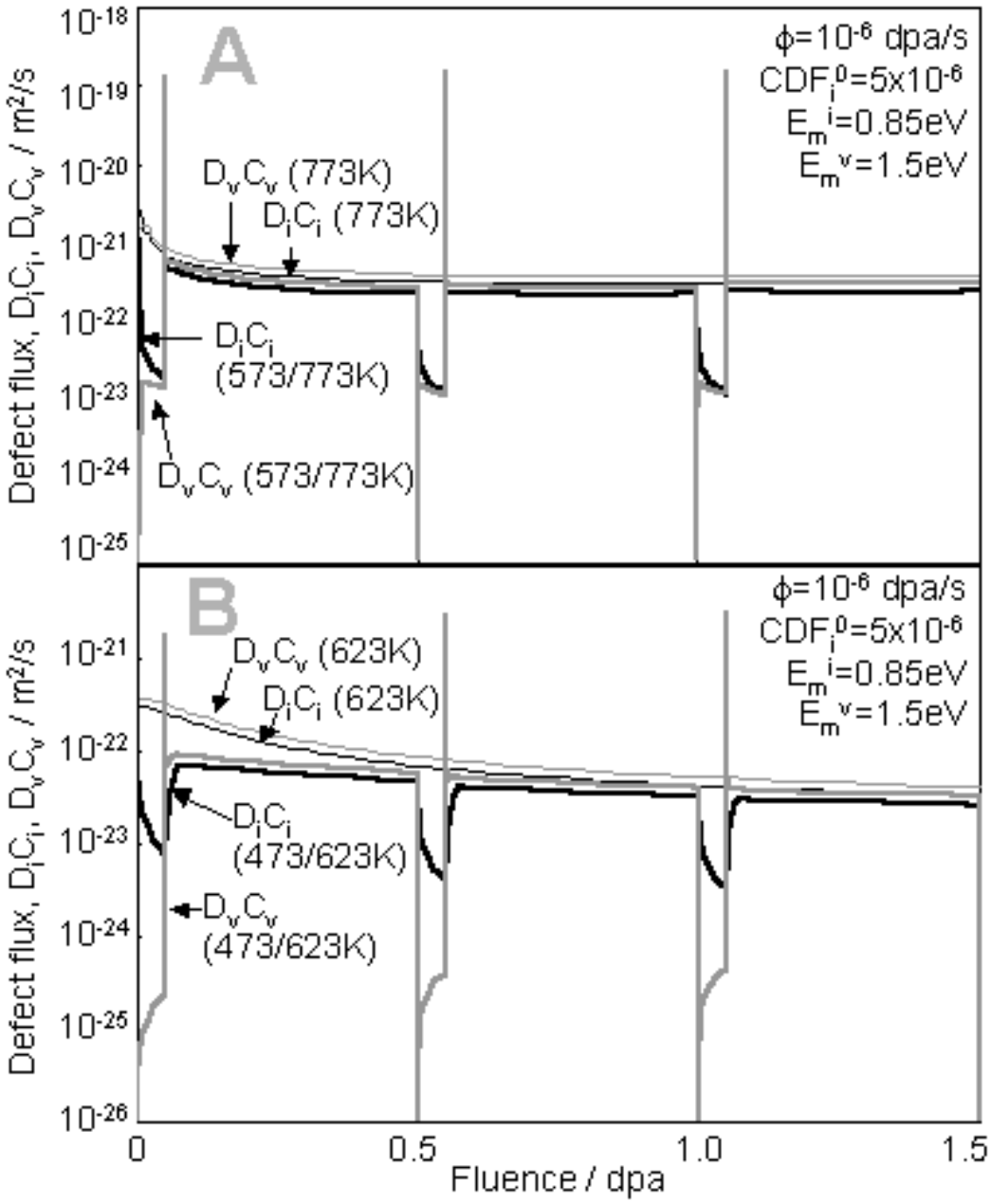
Investigations of radiation-induced microstructural evolution: Current results, status, and planned using from rate theory

- **Most extensive development of rate theory models took place between late 1970s and 1980s**
  - -- demonstrated primary role of radiation-induced microstructural evolution in void swelling and irradiation creep, co-evolution of dislocation structure and cavities
  - -- importance point defect and helium partitioning
  - -- highlighted role of helium and residual gases in driving void swelling by promoting conversion of gas-stabilized bubbles into voids
  - -- good agreement between model predictions and a broad range of data was obtained
  - -- model limitations compensated for by use of “effective” material parameters in some cases
- **Recent and ongoing activities follow from:**
  - -- use of models to predict behavior in irradiation special purpose irradiation experiments
  - -- results of atomistic calculations reveal physical mechanisms not accounted for in earlier models, notably high fraction of in-cascade interstitial clustering and interstitial cluster mobility
  - -- computational advances permit development and application of more detailed kinetic models
  - -- issues related to effects of transmutant helium, helium-vacancy clustering, helium partitioning

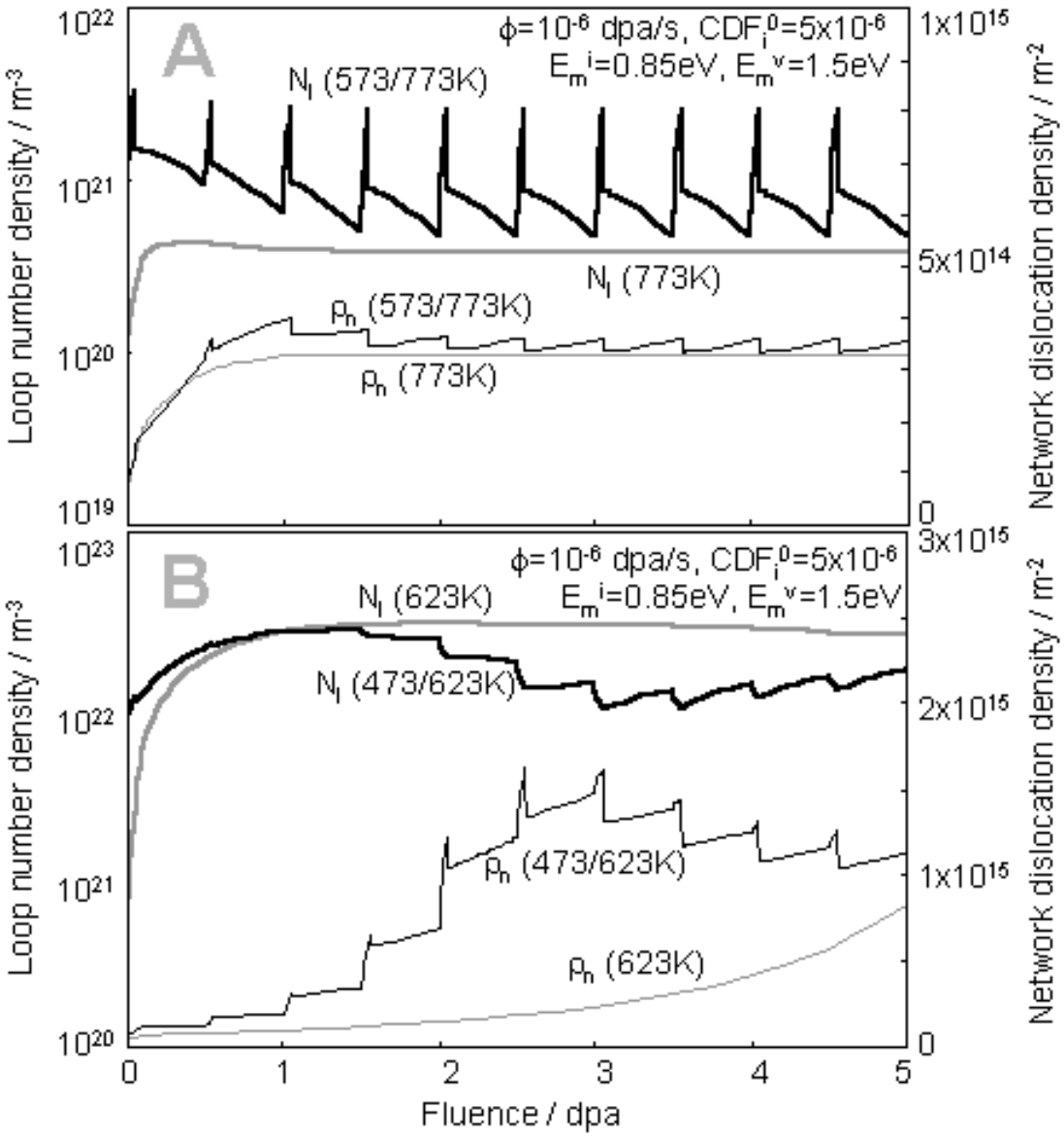
Simulations of Fusion Materials Irradiation Experiment HFIR-RB-13J: Varying Temperature Experiment

- experiment designed to investigate the impact of temperature variations on microstructural evolution, simulate effect of reactor duty cycle
- constant-temperature capsules at 350 and 520˚C and capsules in which the temperature was varied between 220 and 350˚C, and between 350 and 520˚C
- 10% of dose accumulated at the lower temperature at the start of each HFIR cycle, total of 8 cycles, ~5 dpa
- previously developed model modified in a simple way to account for in-cascade clustering of interstitials and interstitial cluster mobility, improved agreement with loop density data at T<400˚

Varying the irradiation temperature leads to successive transients in the point defect concentrations



Point defect transients lead to increased nucleation of small interstitial loops and thereby higher network dislocation densities



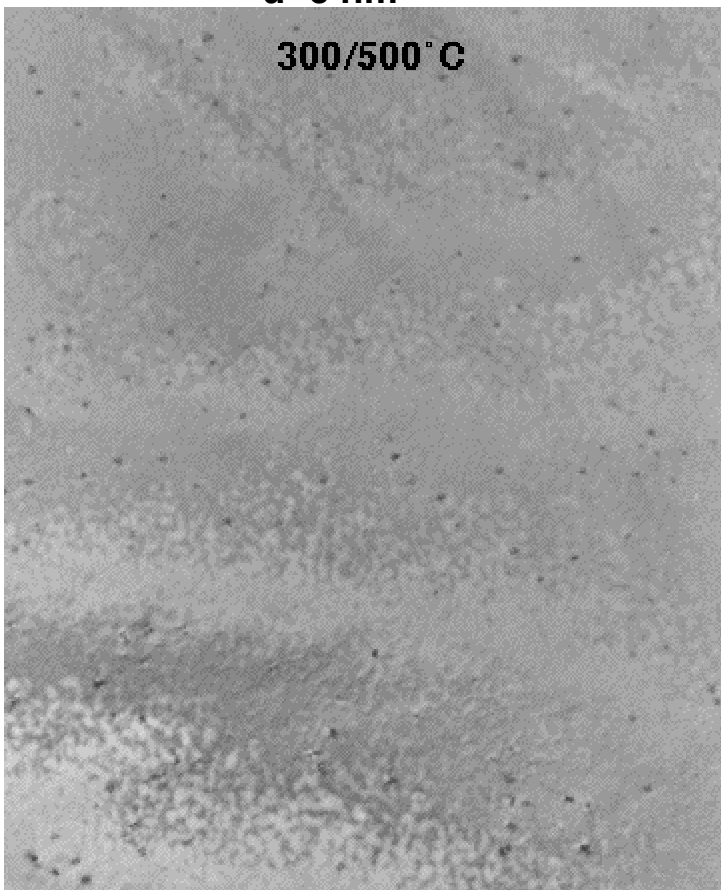
Overall effect is modest, predictions generally consistent with limited data currently available from experiment

Stainless Steel Under Constant 500°C and Varying 300/500°C Irradiation

Periodic irradiation at 300°C leads to enhanced growth of small defect clusters

$\underline{N}$~3x10^{21} m^{-3}
$\overline{d}$=2.5 nm

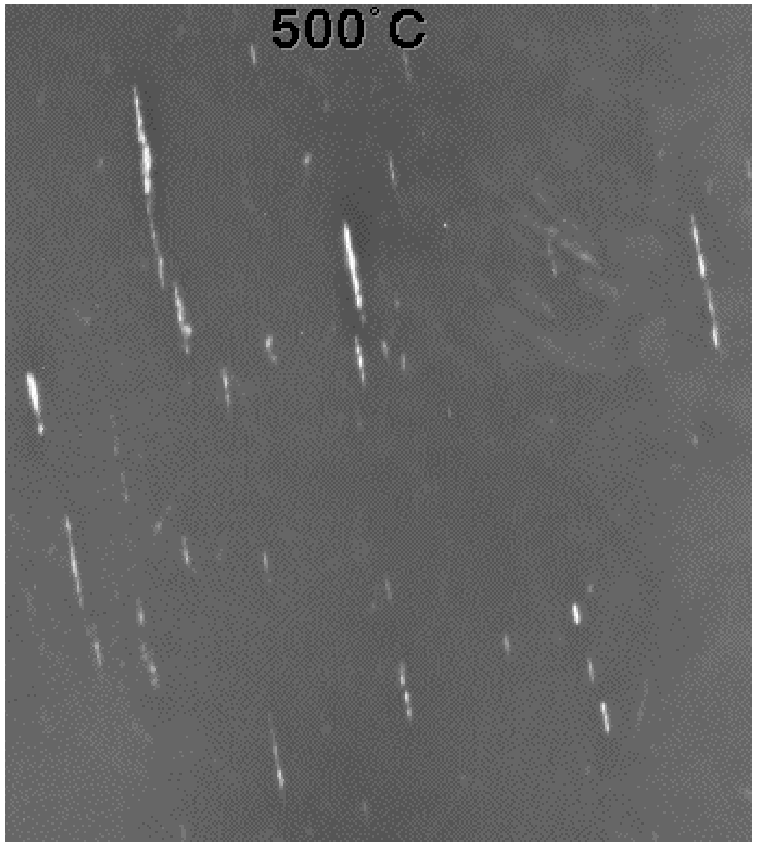
$\underline{N}$~3x10^{21} m^{-3}
$\overline{d}$=6 nm



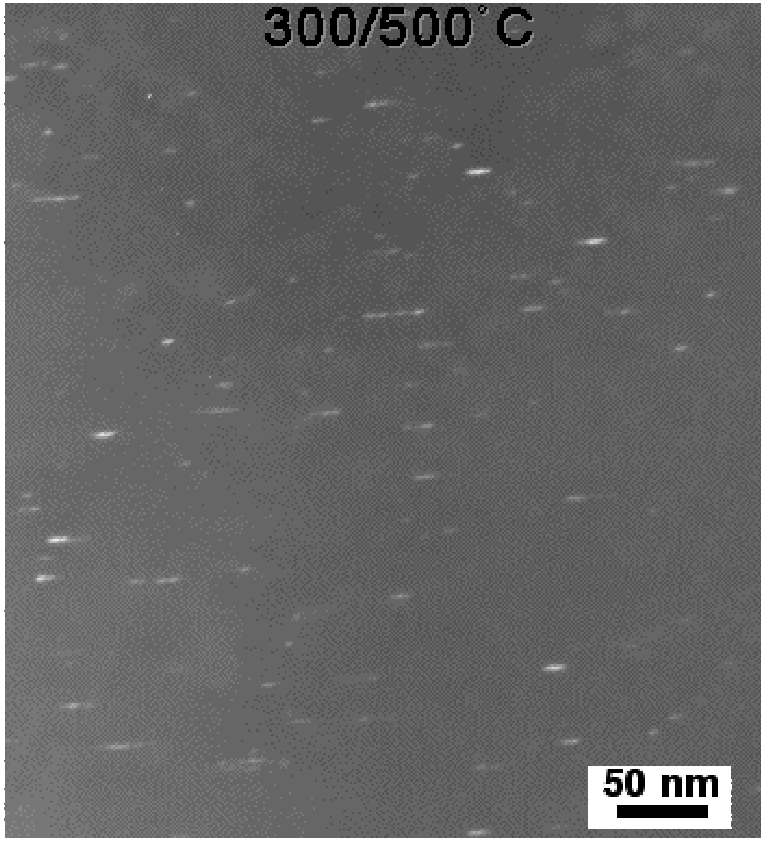
V-4Cr-4Ti Under Constant 500˚C and Varying 300/500˚C Irradiation

Periodic irradiation at 300˚C produces higher density of smaller precipitates

$\underline{N}$~1.2x10^{22} m^{-3}
$\overline{d}$=24 nm



$\underline{N}$~1.2x10^{22} m^{-3}
$\overline{d}$=11 nm



Helium-effects issues relevant to current fusion program needs

- **Helium diffusion and transport mechanisms**
- **Helium trapping and partitioning between different microstructural sinks**
  - -- Bubble nucleation in matrix, on precipitates and grain boundaries
  - -- Helium flow to grain boundaries
- **Bubble growth and evolution of size distribution, swelling**
- **Grain boundary bubble growth under stress**
- **Creep rupture**

Bubble Growth & Size Evolution in Steels

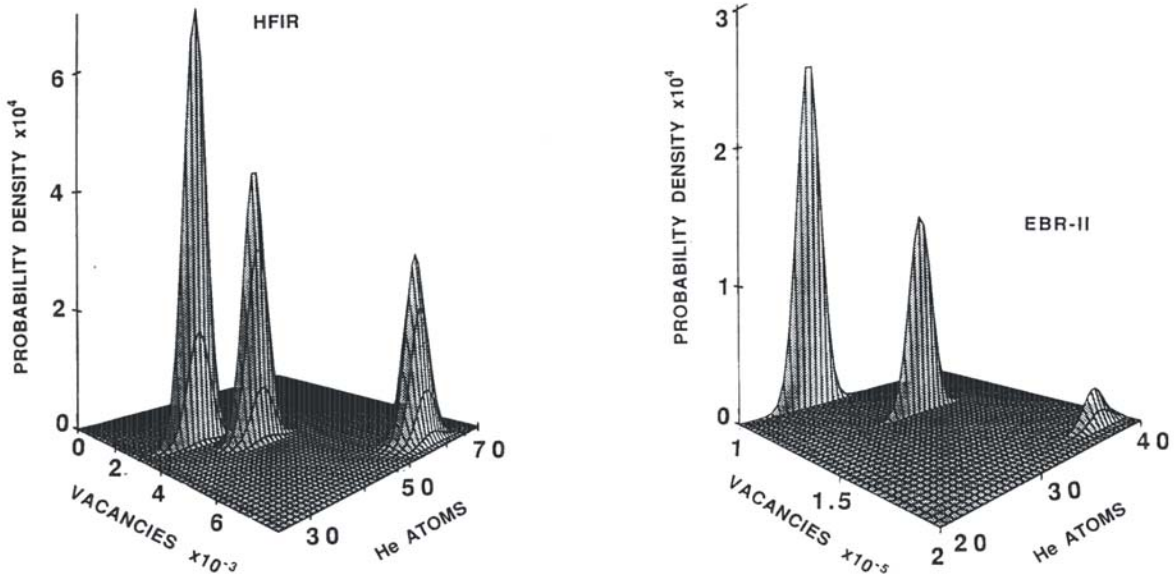


Fig. (2): Computed non-equilibrium vacancy-helium cluster size distributions under low (left), and high (right) helium generation rates.

Modeling Helium Bubbles in Vanadium

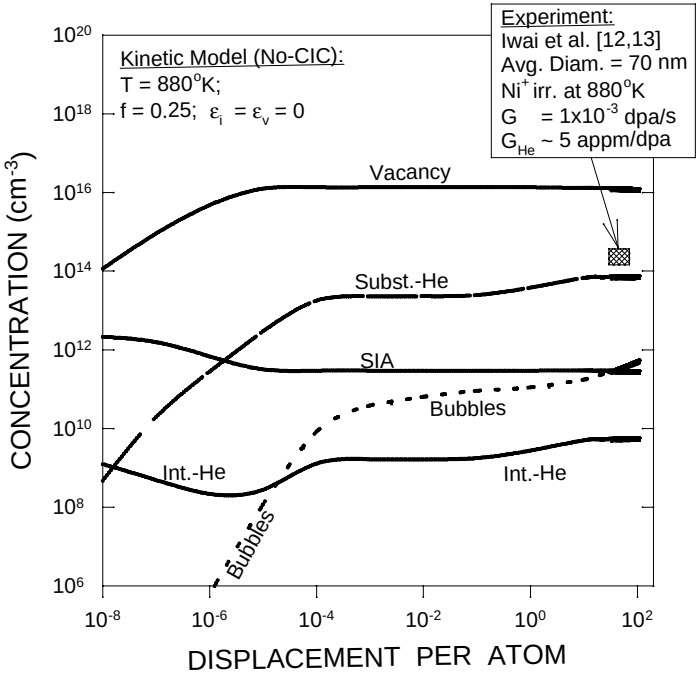


Figure 1: Without CIC formation: ***$N_{ppt} = 10^{13} cm^{-3}$, $R_{ppt} = 30$ nm, grain-size = 3μm, dislocation density = 10^9 cm^{-2}, T = 880 K, G = 1x10-3 dpa/s, G_{He}=5 appm/dpa, f = 0.25, $\varepsilon_i = \varepsilon_v = 0$).***

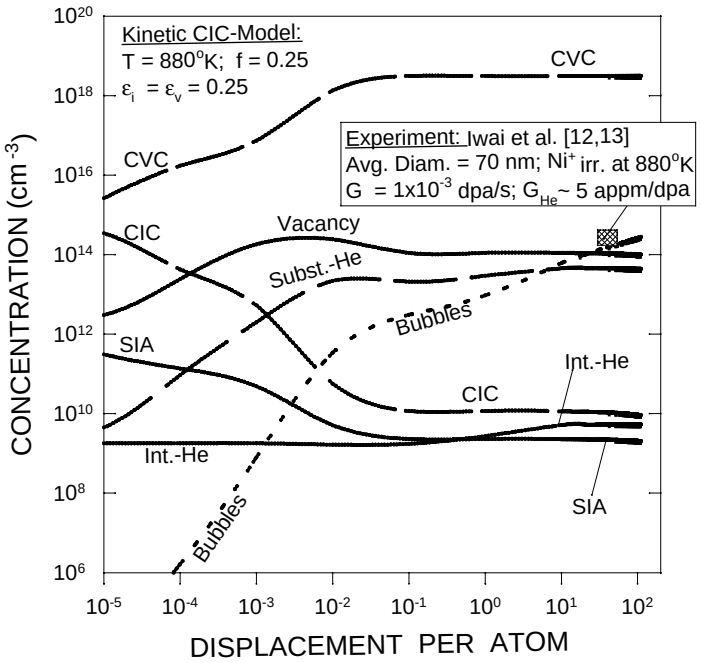
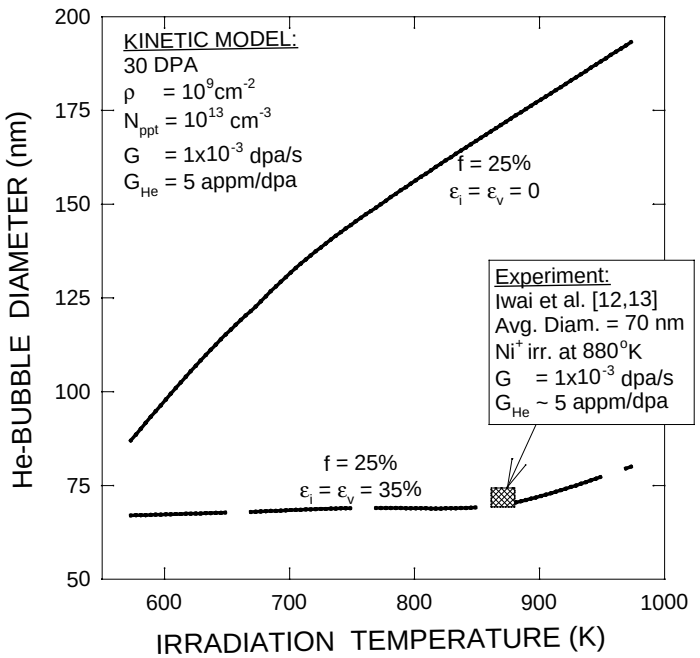
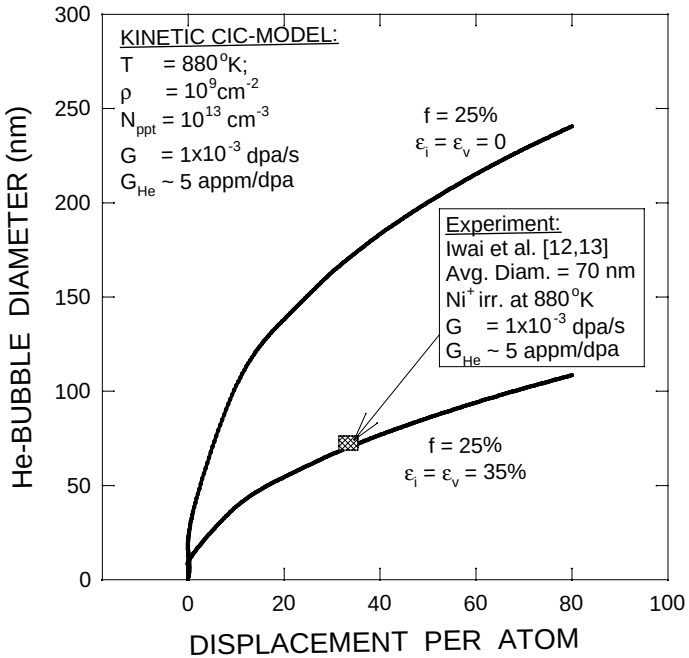


Figure 2: With CIC formation

Modeling Helium Bubbles in Vanadium - 2





Summary of Rate Theory Work

- **Current models are generally successful at predicting radiation-induced microstructural evolution, e.g. temperature and dose dependence of void swelling in austenitic stainless steels**
- **Additional model development focused on:**
  - -- expand range of materials, requires developing microstructural database for model verification
  - -- incorporating new physical damage production mechanisms observed in MD simulations (while validity of these mechanisms are being examined)
  - -- evaluating impact of MD and MC-based primary damage parameters
  - -- exploit new computational tools, e.g. by developing more detailed defect clustering (i.e. point defects, solutes, He, ...) models to improve description of nucleation
  - -- develop models to account for solute and precipitate effects more explicitly, e.g. for application to ODS ferritic alloys
  - -- improved description of helium partitioning, obtain guidance from atomistic simulations
- **Interact with experimentalists to evaluate both models and experiments, e.g. varying-temperature experiment**

Summary and Future Work

- **Hierarchical multiscale-multiphysics approach required to simulate radiation effects on fusion reactor materials**
  - -- phenomena span atom dimensions to meters, femtoseconds to decades
  - -- primary damage production and gas generation
  - -- damage diffusion and accumulation (cascade aging) produces nanostructural and nanochemical features
  - -- microstructural changes lead to changes in mechanical properties, e.g. radiation hardening and flow localization
- **Fusion materials program possesses or is developing modeling tools required to treat each of the closely coupled phenomena**
- **Current and future research: develop and apply integrated, MSMP modeling techniques to fusion-relevant materials in the fusion environment**
  - -- high displacement damage rates
  - -- high gas (He, H) generation rates