Computational Modeling of Defects and Microstructure Dynamics in Materials under Irradiation

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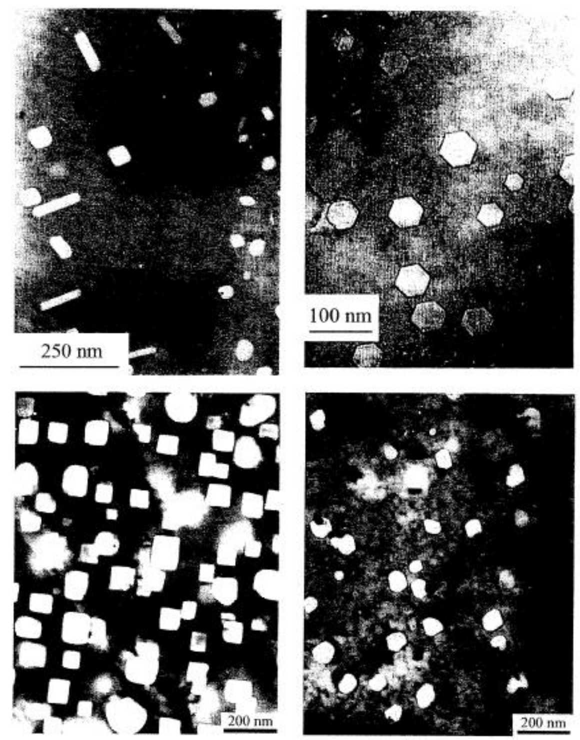
Motivation

- Irradiation damage results in complex processes of microstructural and compositional changes in materials
- These processes are all driven by production, diffusion and reactions of point defects
- Classical modeling approaches (e.g., clustering and nucleation theory, rate theory) are not adequate

Void formation

- Caused by vacancy super-saturation
- It can be homogeneous or heterogeneous
- Shape depends on crystal type
- Void lattice is possible
- Coupling with stress and compositional changes
- In the presence of gas atoms, voids turn into gas bubbles



Irradiation-induced voids in
(a) steel, (b) aluminum and (c) & (d) magnesium

Research objective

- Develop a unified **mesoscale** model to predict the **concurrent** microstructural and compositional changes in irradiated materials

- **Mesoscale →** resolve space

- **Concurrent →** processes are all driven by point defects generated by irradiation

- Materials systems under consideration:

  - Pure metals
  - Metallic alloys
  - Oxides

  without and with gas in the matrix

Why the mesoscale?

Breakthroughs in understanding and predicting materials performance can be made through success at the mesoscale because this is where the materials complexity reveals itself; the mesoscale materials models fold the fundamental materials properties with the microstructural complexity to both predict and understand the macroscopic response of materials …

Approach

- Non-equilibrium thermodynamics
- Field theory of defects and microstructure
  → **phase field** theory
- Statistical physics underpinning

Typical phase-field models

A typical phase field model is developed in two steps:

- Construct a free energy functional of the system
- Derive kinetic equations following Onsager formalism of non-equilibrium T.D.

$$F[c,\eta] = \int_{\Omega} f(c,\eta)\, d\Omega$$

$$\frac{\partial c}{\partial t} = \nabla \cdot M \nabla \frac{\delta F}{\delta c} + \xi(\mathbf{x}, t)$$

Cahn-Hilliard Eq.

$$\frac{\partial \eta}{\partial t} = -L \frac{\delta F}{\delta \eta} + \zeta(\mathbf{x}, t)$$

Allen-Cahn (G.L.) Eq.

Conservation properties

For a system decaying towards a lower energy state, the last kinetic equations satisfy two conditions:

- Free energy decay (irreversibility)

$$\frac{d}{dt}F[c,\eta] = \frac{d}{dt}\int_{\Omega} f(c,\eta)\, d\Omega \le 0$$

- Mass conservation

$$\frac{d}{dt}M[c,\eta] = \frac{d}{dt}\int_{\Omega} m(c,\eta)\, d\Omega = 0$$

Conservation properties under irradiation

Irradiated materials are *driven systems*; irradiation deposits energy and "mass" into the system

- Free energy is not necessarily decreasing with time …

$$\frac{d}{dt}F[c,\eta] = \frac{d}{dt}\int_{\Omega} f(c,\eta)\, d\Omega \quad ?$$

- Mass is not necessarily constant …

$$\frac{d}{dt}M[c,\eta] = \frac{d}{dt}\int_{\Omega} m(c,\eta)\, d\Omega \quad ?$$

- Mass $\rightarrow$ conserved order parameters (defects or actual atoms)

Phase-field model for irradiated materials

Follow same steps without irradiation and add sources to account for generation and reactions.

$$F[c,\eta] = \int_{\Omega} f(c,\eta)\, d\Omega$$

no irradiation

modified Cahn-Hilliard Eq.

$$\frac{\partial c}{\partial t} = \nabla \cdot M \nabla \frac{\delta F}{\delta c} + \xi(\mathbf{x},t) + G(\mathbf{x},t) - R(\mathbf{x},t)$$

modified Allen-Cahn (G.L.) Eq.

$$\frac{\partial \eta}{\partial t} = -L \frac{\delta F}{\delta \eta} + \zeta(\mathbf{x},t) + \zeta_{Irrad}(\mathbf{x},t)$$

This is formally equivalent to replacing *F* (c,η) with a Lyapunov functional *J* (c,η) and using the latter to derive governing eqns.

Example: void formation due to vacancy supersaturation

$$F[c_v,\eta] = \int_\Omega \Big[f_o^m(c_v)$$

Energy of a matrix with point defects

$$+ \quad w(c_v,\eta)$$

Landau energy term (bi-stability: matrix phase versus void phase)

$$+ \quad \kappa_v \left|\nabla c_v\right|^2 + \quad \kappa_\eta \left|\nabla \eta\right|^2$$

Gradient terms due to field inhomogeneity

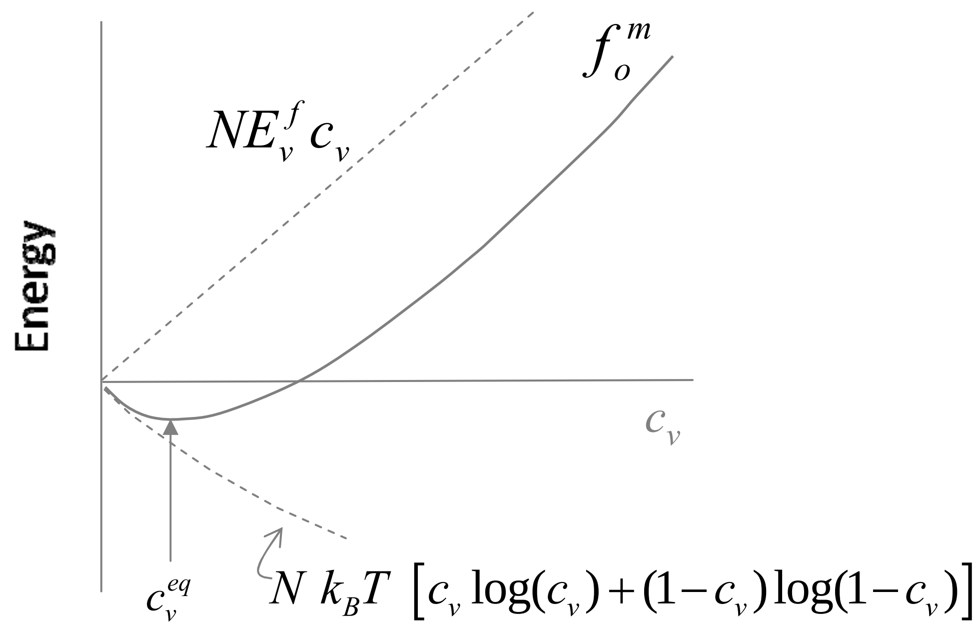
$$+ \quad f^{\,elastic} \Big] \, d\Omega$$

Stress-defect interaction energy

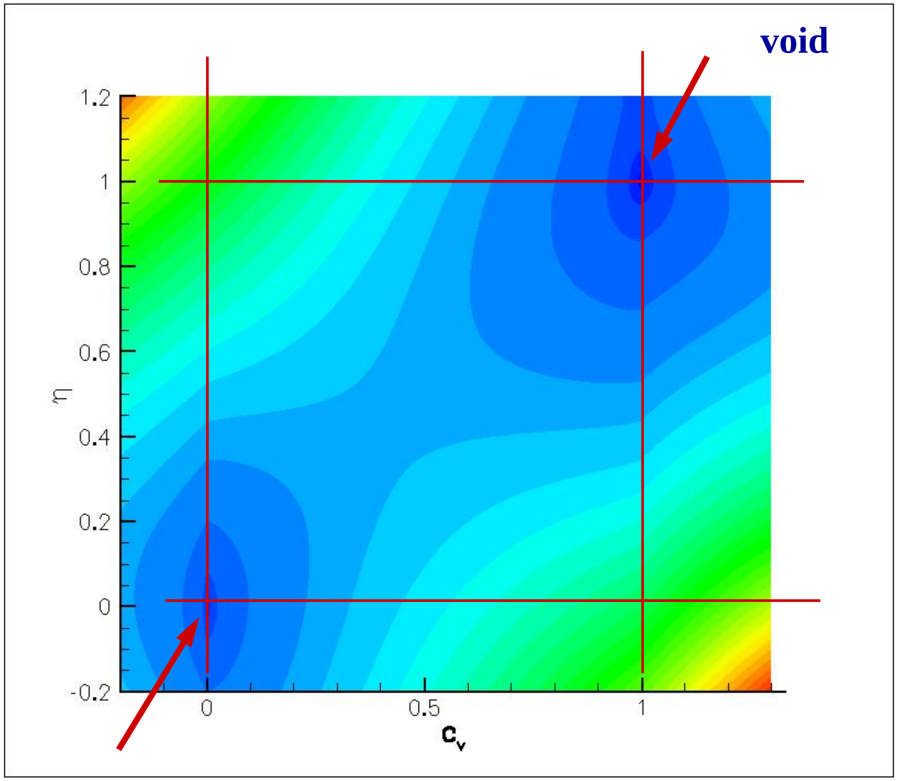
Point defect energy in matrix

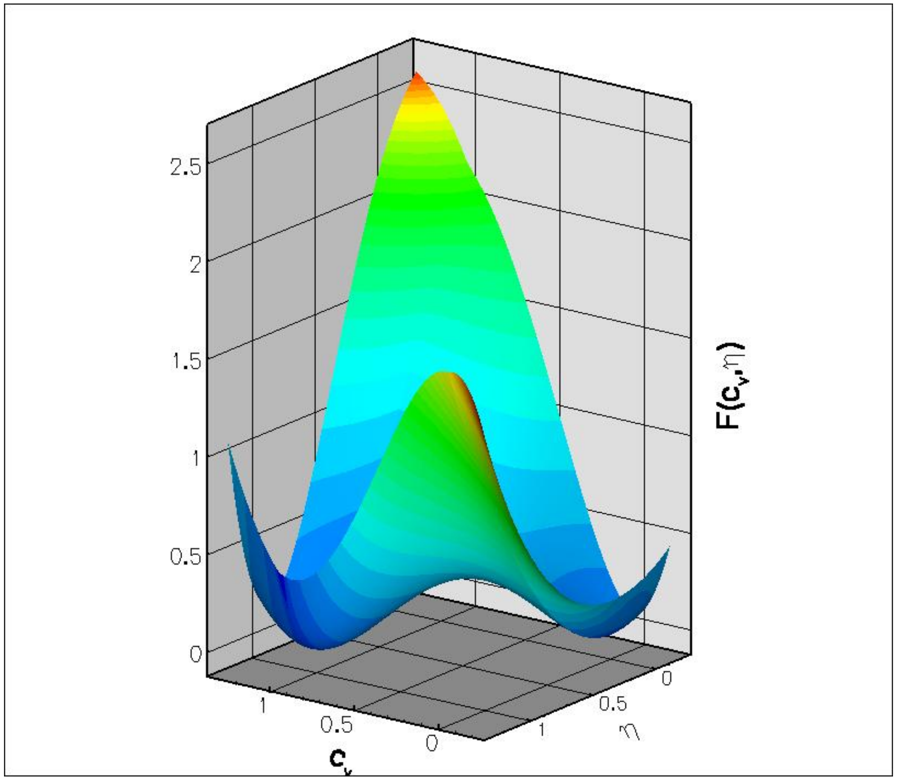
$$f_o^m = N\,E_v^f c_v \quad + N\,k_B T\left[c_v \log(c_v) + (1-c_v)\log(1-c_v)\right]$$

Enthalpic + Entropic energy terms



Energy landscape for matrix with vacancies



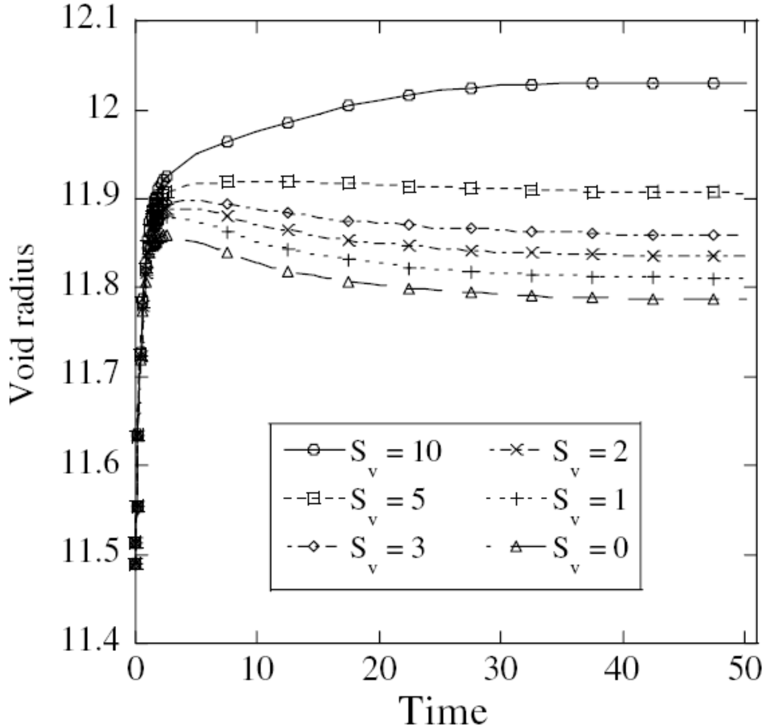


matrix with thermal equilibrium concentration

Numerical tests

- **Void growth and shrinkage (Gibbs-Thompson Effect)**
- **Interaction between voids (Ostwald ripening)**
- **Nucleation of voids (homogeneous)**
- **Nucleation in the vicinity of pre-existing void**

Void growth and shrinkage

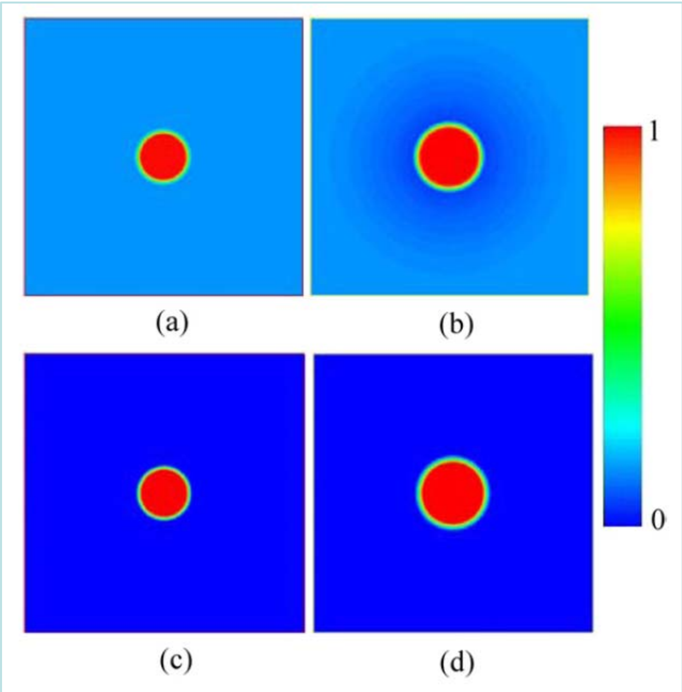


Growth and shrinkage take place depending the background concentration and the void radius

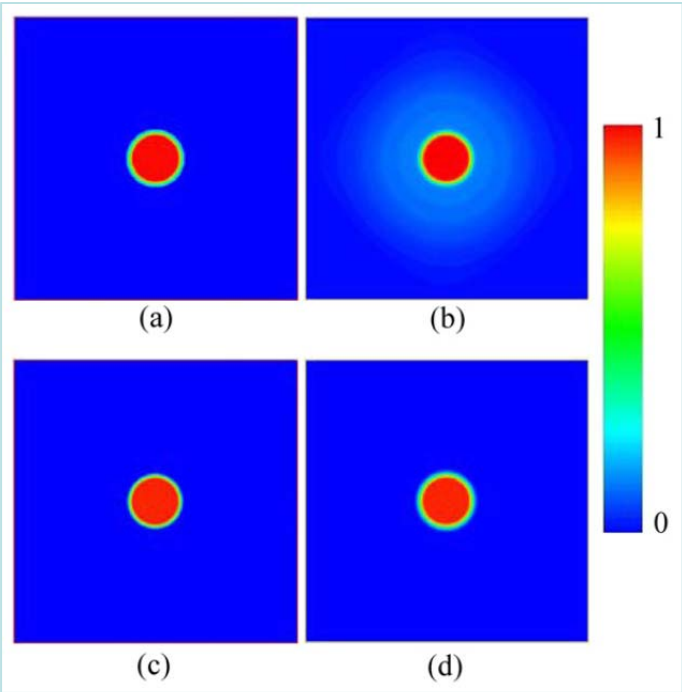
Gibbs-Thompson Effect

Figure 5. Void radius as a function of time for different initial vacancy supersaturation levels.

Void growth and shrinkage



growth

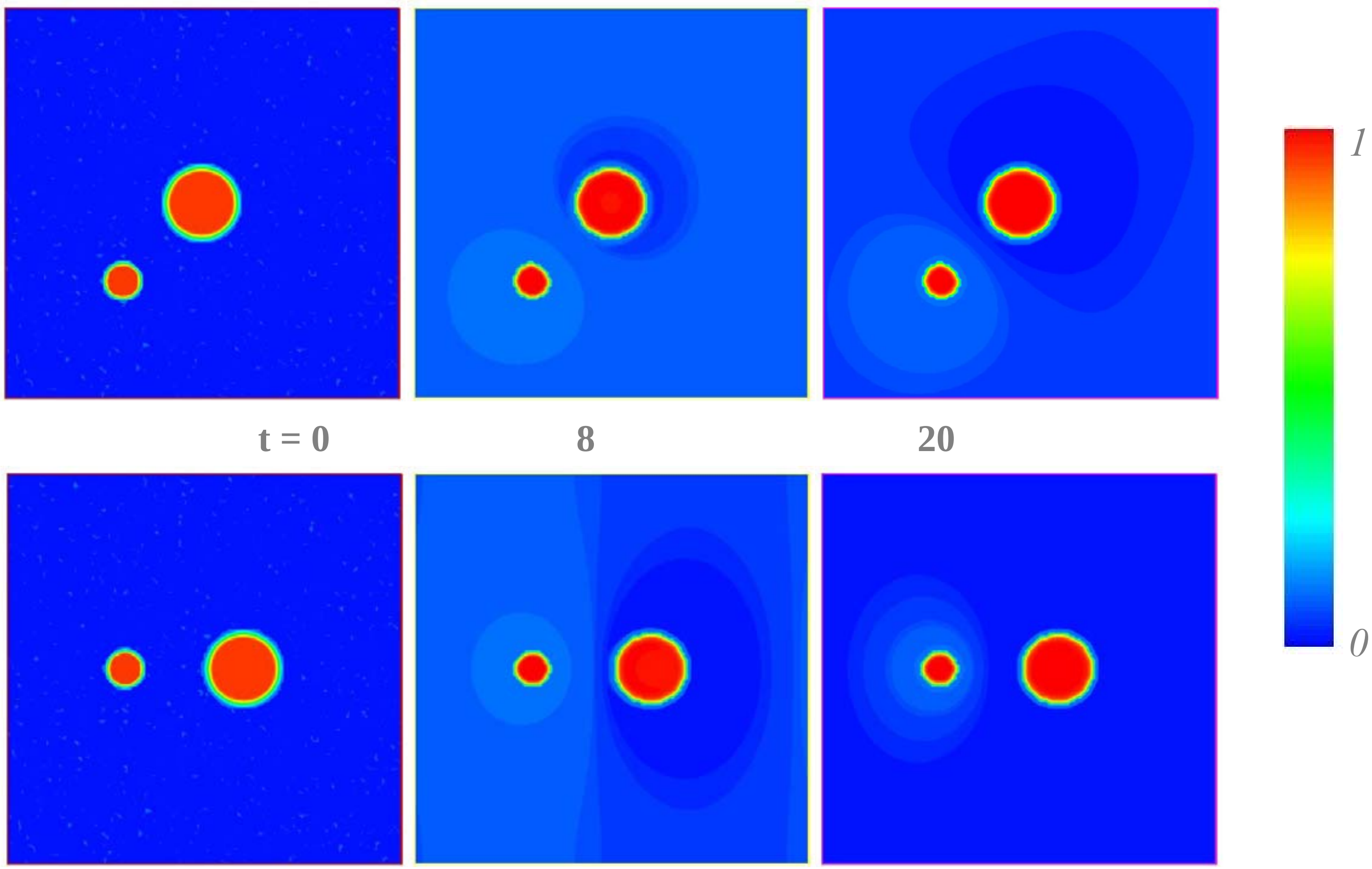


shrinkage

Void-void interaction

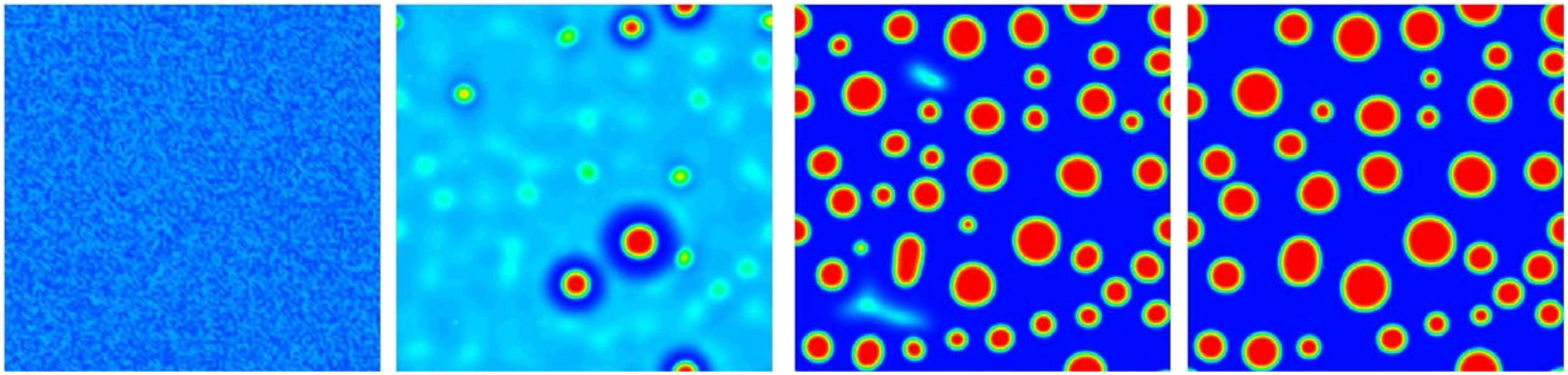
Ostwald ripening example

Large voids grow at the expense of small ones



Interaction between two voids surrounded by unsaturated matrix, r_1=5, r_2 = 10

Homogeneous nucleation of voids under vacancy generation

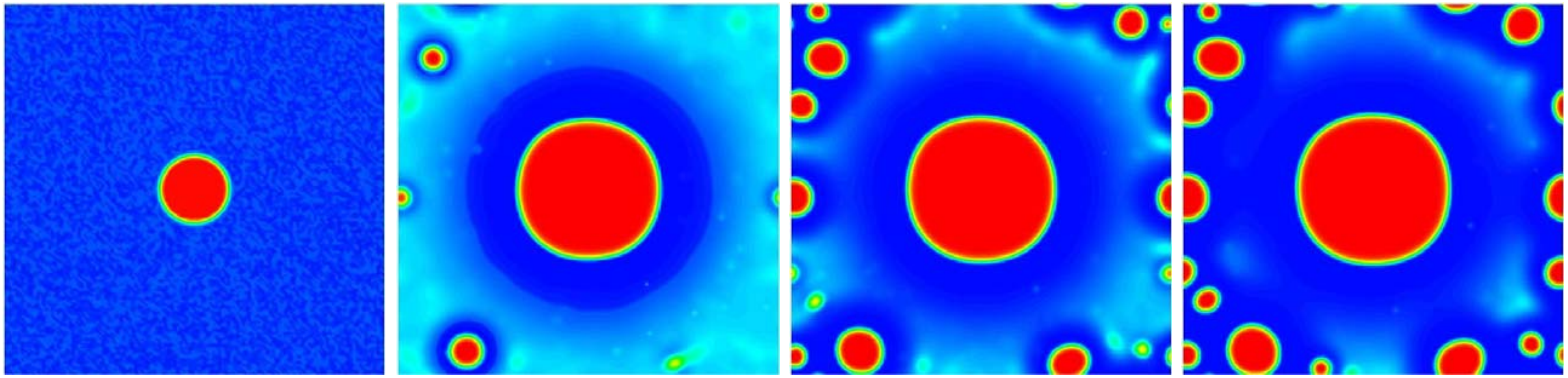


t = 0   165   190   250

Vacancy field evolution showing void nucleation due to radiation induced vacancies

Voids nucleate due to fluctuations in the vacancy concentration field. The nucleation process is homogeneous

Nucleation close to a pre-existing void



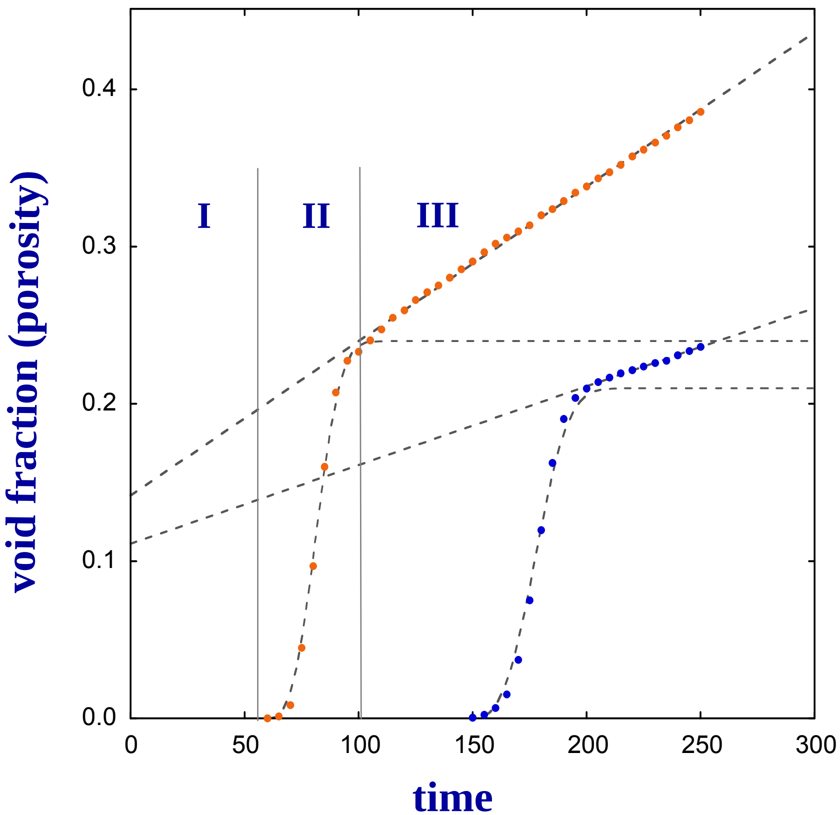
t = 0 210 240 250

Vacancy field evolution showing void growth in the presence of radiation effects

Initial void grows while new voids nucleate …

Ripening suppresses the small voids nucleating in the vicinity of the large one.

Analysis of nucleation and growth



Stage I : Incubation period

Stage II: Nucleation regime
(Johnson-Mehl-Avrami Equation)

$$p = p^e\left(1 - \exp\left(-kt^3\right)\right)$$

$p^e = 0.24, k = 6.73\times10^{-5}$

$p^e = 0.21, k = 2.88\times10^{-5}$

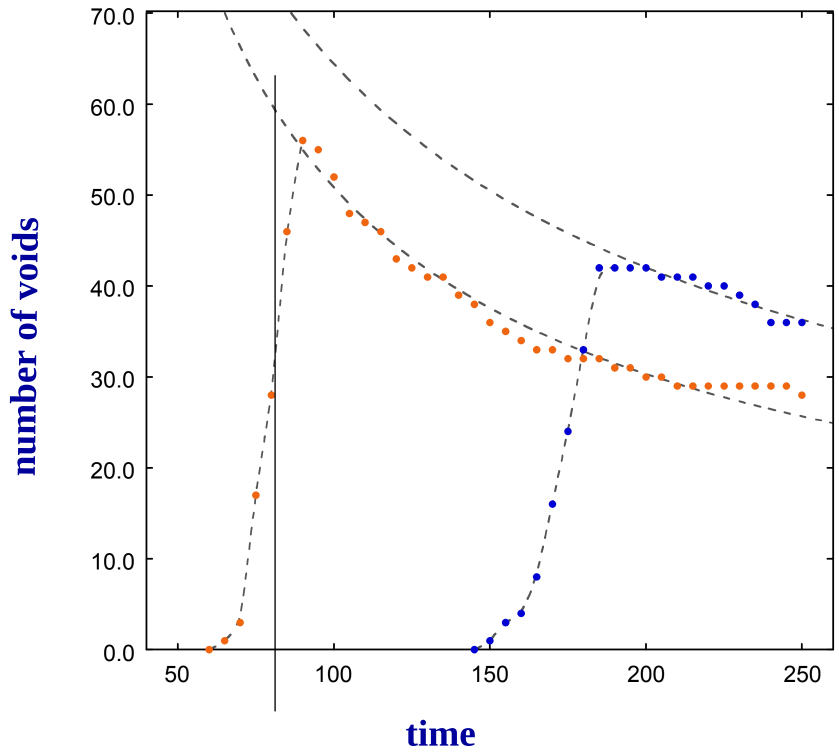
Stage III: Growth regime
(Ostwald ripening)

$$p = p^o\left(1 + t/\tau\right)^1$$

$p^o = 0.14185, \tau = 144.61$

$p^o = 0.11119, \tau = 222.51$

Void density as a function of time



Stage II: Nucleation regime

$$N = Jt$$

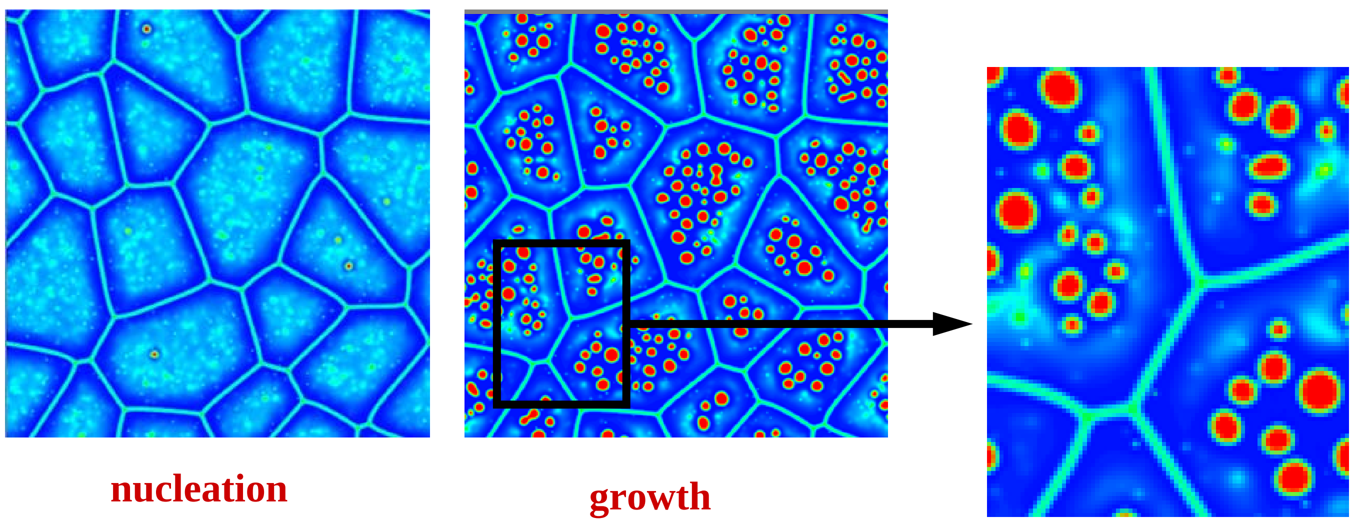
Stage III: Growth (Ostwald ripening)

$$N = N^{o}\left(1 - t/\tau\right)^{-0.75}$$

$N^{o} = 55, \tau = 90.764$

$N^{o} = 45, \tau = 210.34$

Role of grain boundaries

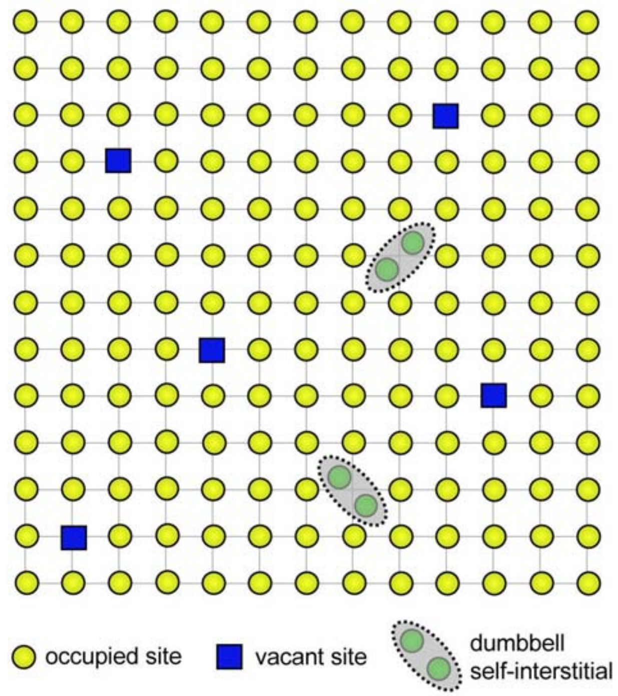


nucleation

growth

denuded GB regions

Introducing interstitials



Phase field model with interstitials included

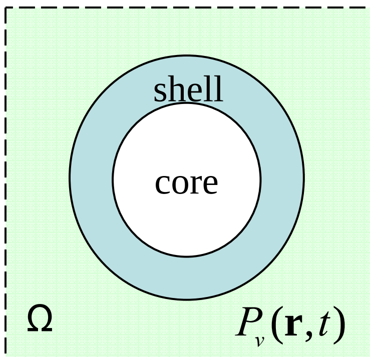
$$F = N\int_V \left[h(\eta) f^s(c_v, c_i) + j(\eta) f^v(c_v, c_i) + \frac{\kappa_v}{2}|\nabla c_v|^2 + \frac{\kappa_i}{2}|\nabla c_i|^2 + \frac{\kappa_\eta}{2}|\nabla \eta|^2 \right] dV$$

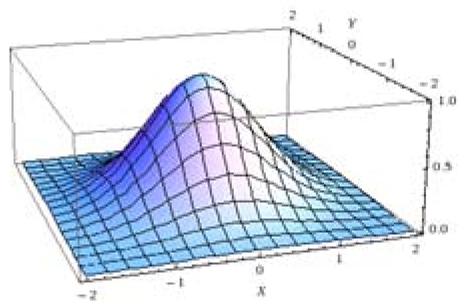
$$\frac{\partial c_v}{\partial t} = \nabla \cdot \left(M_v \nabla \frac{1}{N} \frac{\delta F}{\delta c_v} \right) + \xi(\mathbf{r},t) + P_v(\mathbf{r},t) - R_{iv}(\mathbf{r},t) - S_v^{GB}(\mathbf{r},t)$$

$$\frac{\partial c_i}{\partial t} = \nabla \cdot \left(M_i \nabla \frac{1}{N} \frac{\delta F}{\delta c_i} \right) + \zeta(\mathbf{r},t) + P_i(\mathbf{r},t) - R_{iv}(\mathbf{r},t) - S_i^{GB}(\mathbf{r},t)$$

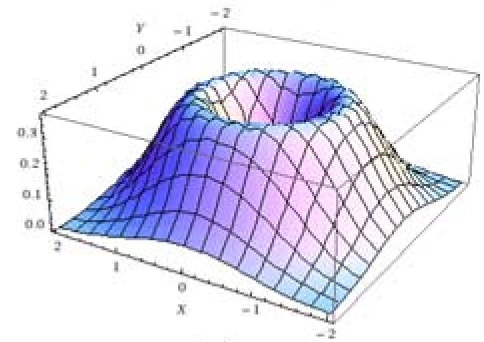
$$\frac{\partial \eta(\mathbf{r},t)}{\partial t} = -L\frac{\delta F}{\delta \eta} + \varsigma(\mathbf{r},t) + P_{v,i}(\mathbf{r},t)$$

Cascade representation



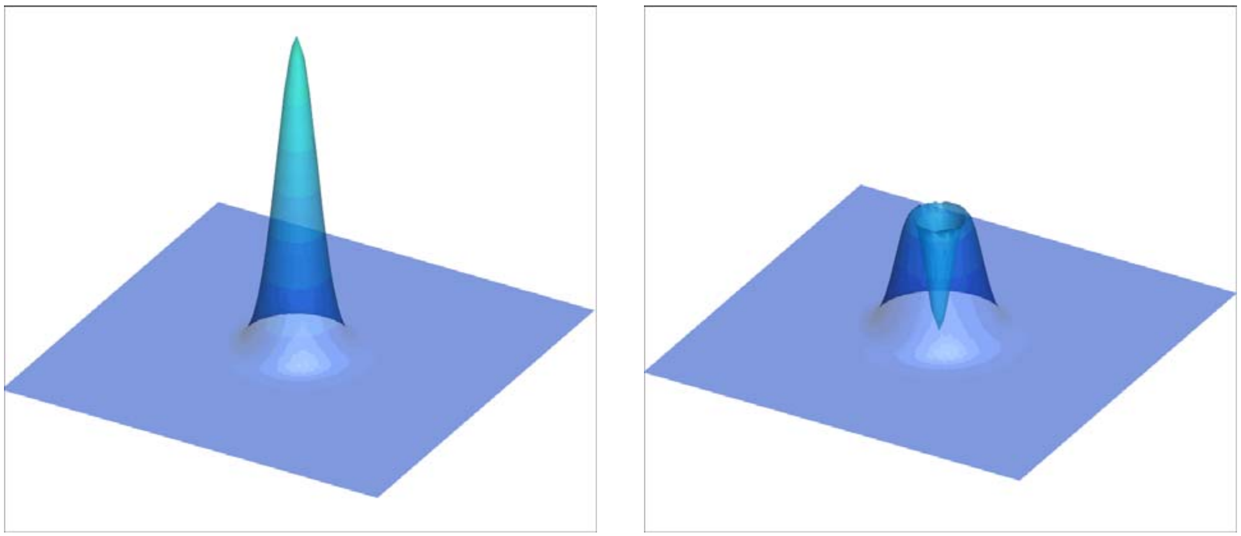


P_v

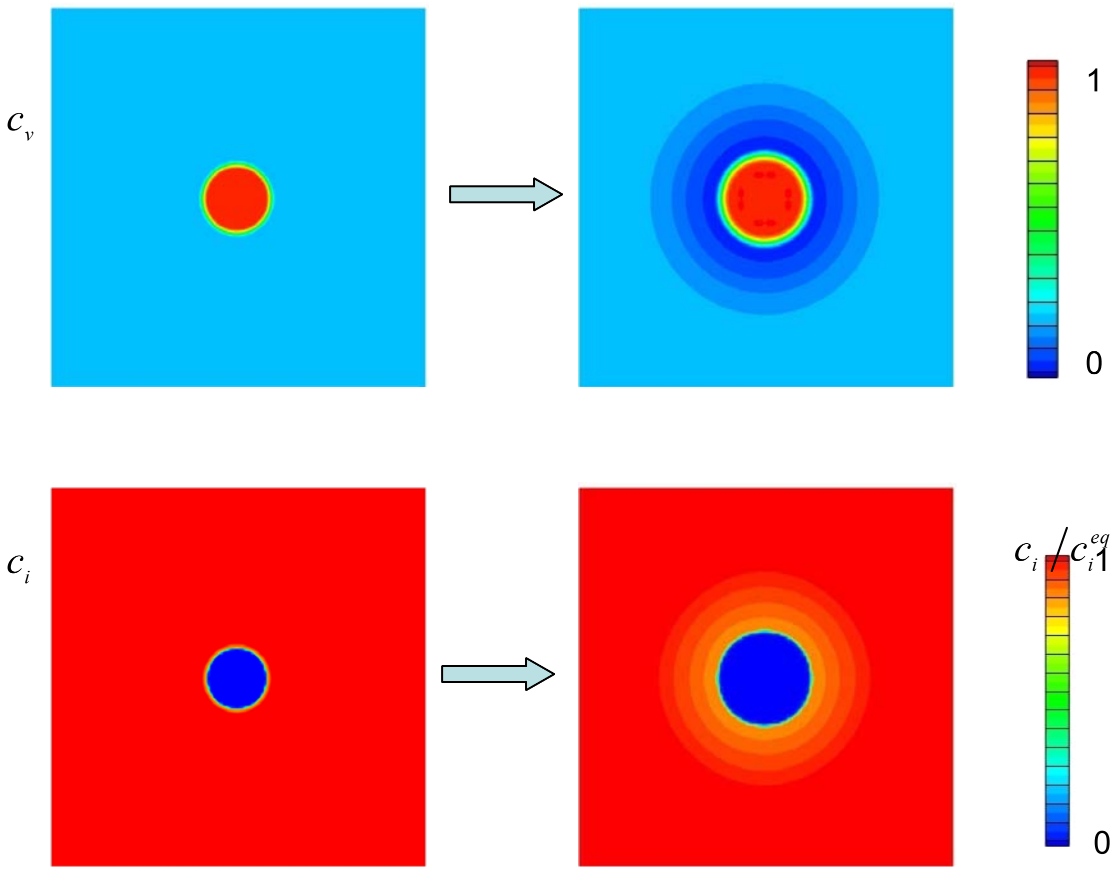


P_i

Evolution of a single cascade
Diffusion and recombination of vacancies and interstitials

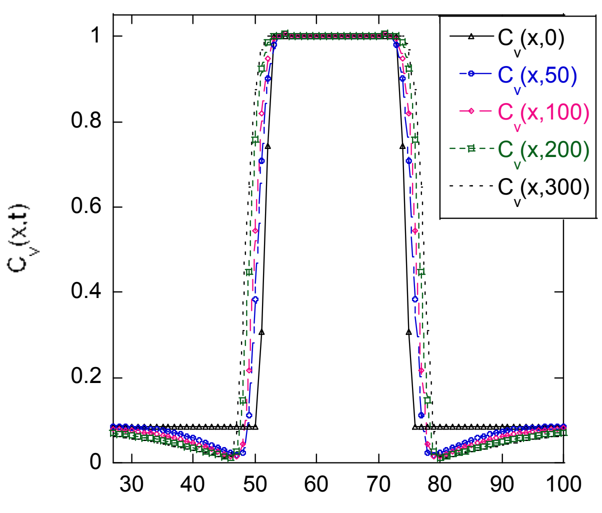


Void Growth



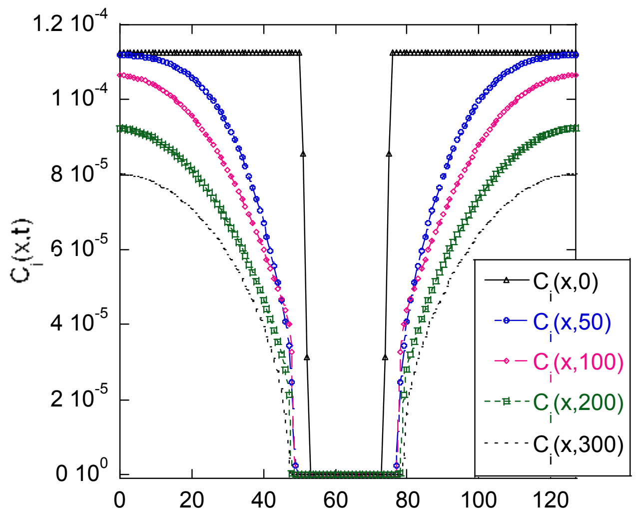
c_v and c_i fields for void growth in the presence of excess vacancies in the surrounding matrix. S_v = 20, S_i = 1.0

Fields profiles during void growth



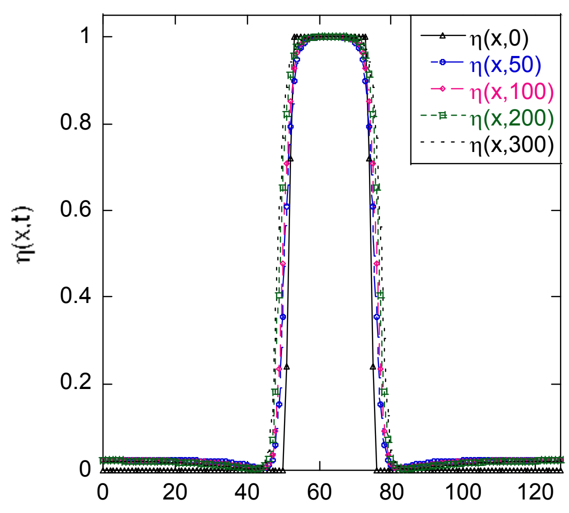
$c_v(x,t)$

Vacancy field



$c_i(x,t)$

Interstitial field

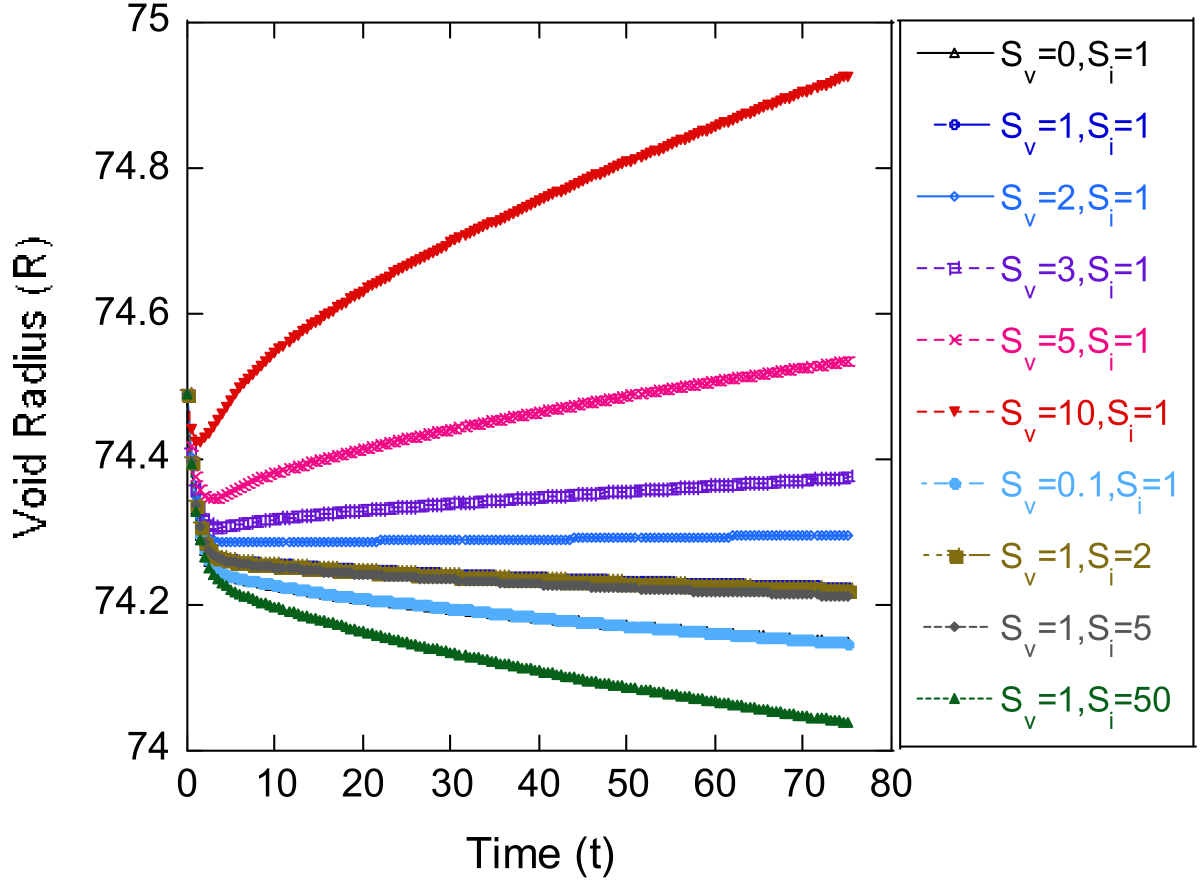


$\eta(x,t)$

Void phase field

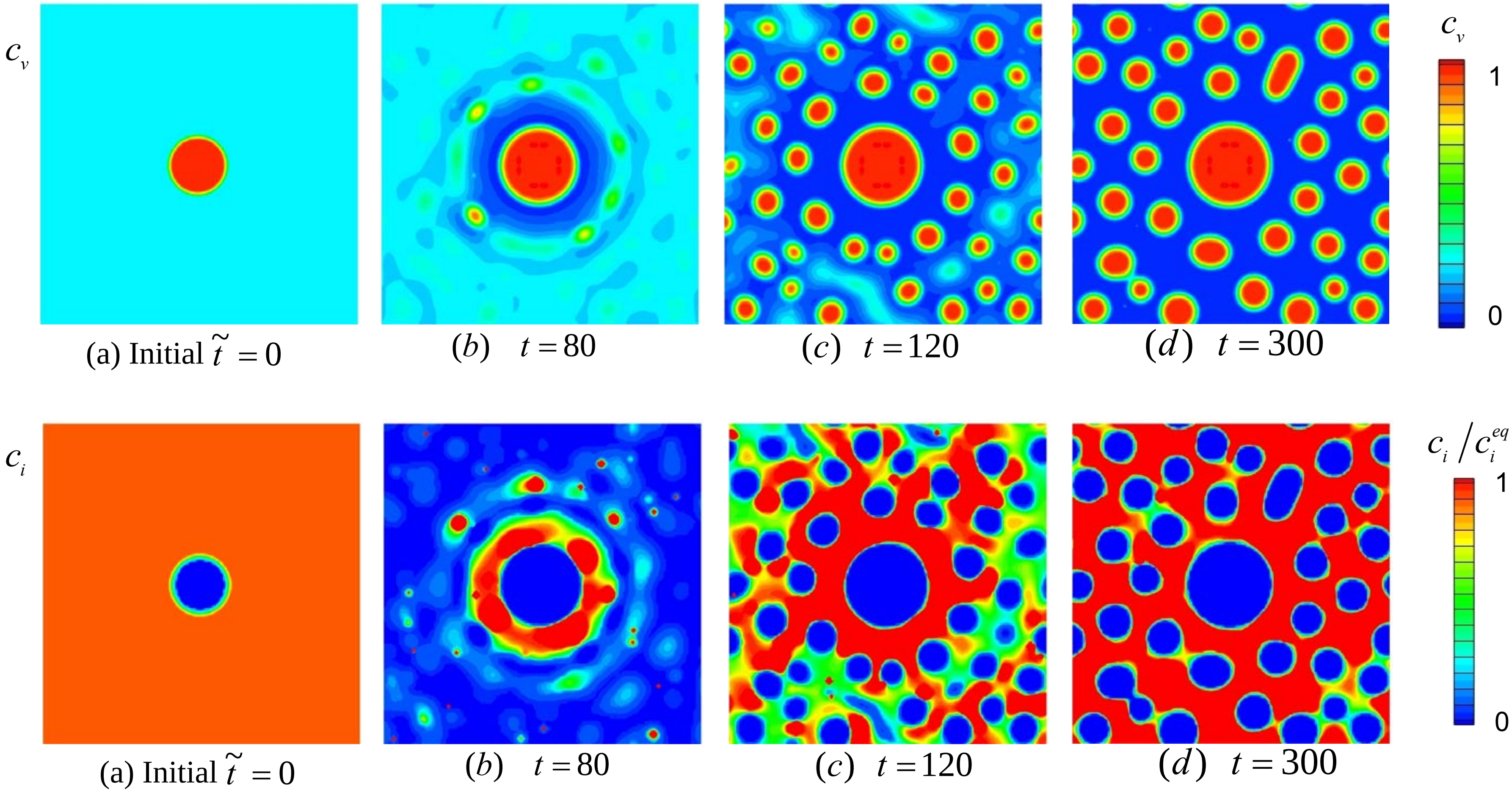
Fields profiles at a cross-section at the center of the simulation cell.
Void growth in the presence of excess vacancies in the surrounding matrix. $S_v = 20$, $S_i = 1.0$ (No radiation source)

Void radius with supersaturation



Void radius as a function of time, for different initial defect supersaturation

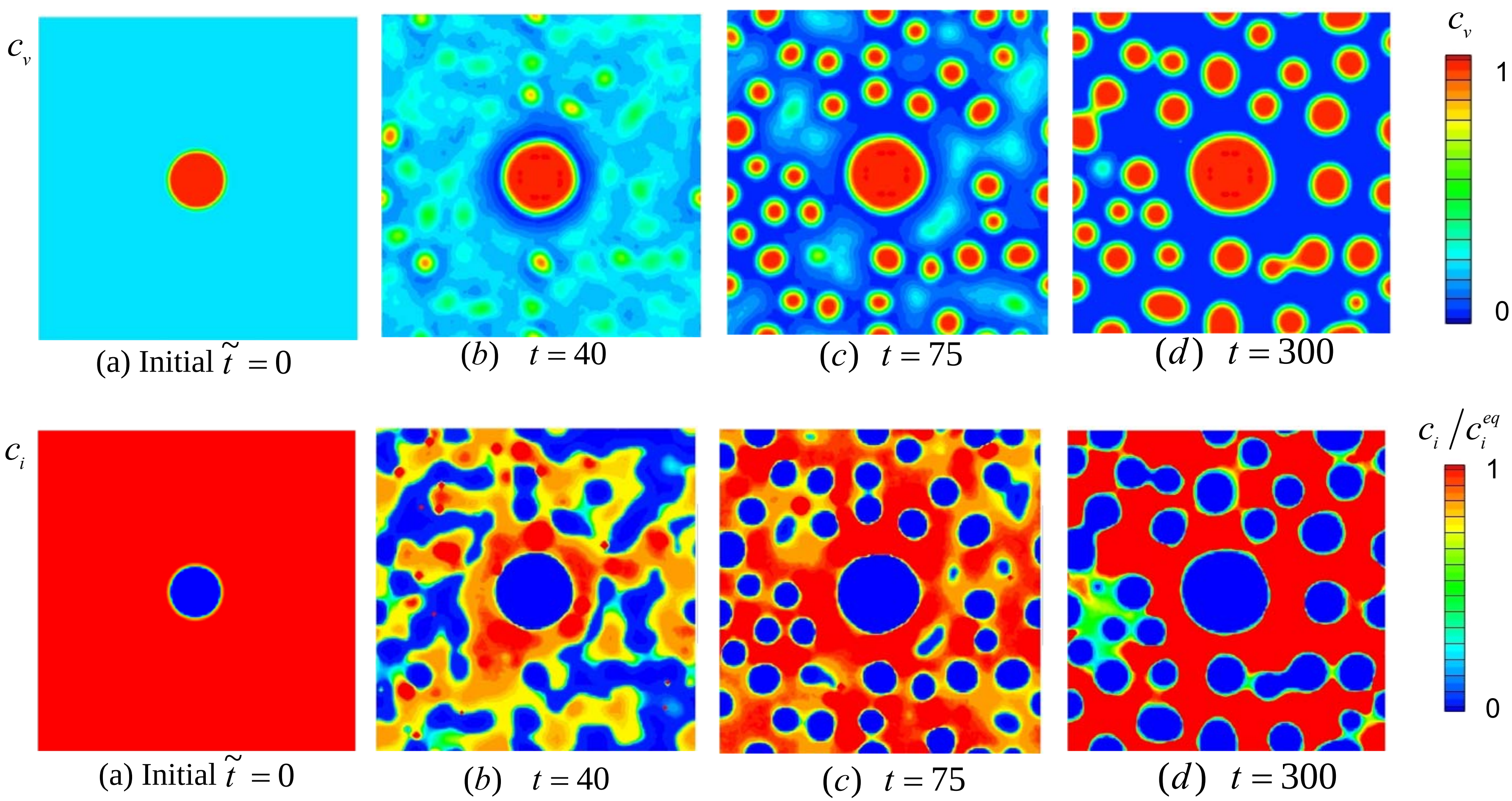
Void growth under irradiation



Vacancy and interstitial field evolution showing void growth in the presence of radiation effects.

S_v=50, S_i = 1, P_v=0.25, P_i = 0.15 (on 128 x 128 grid)

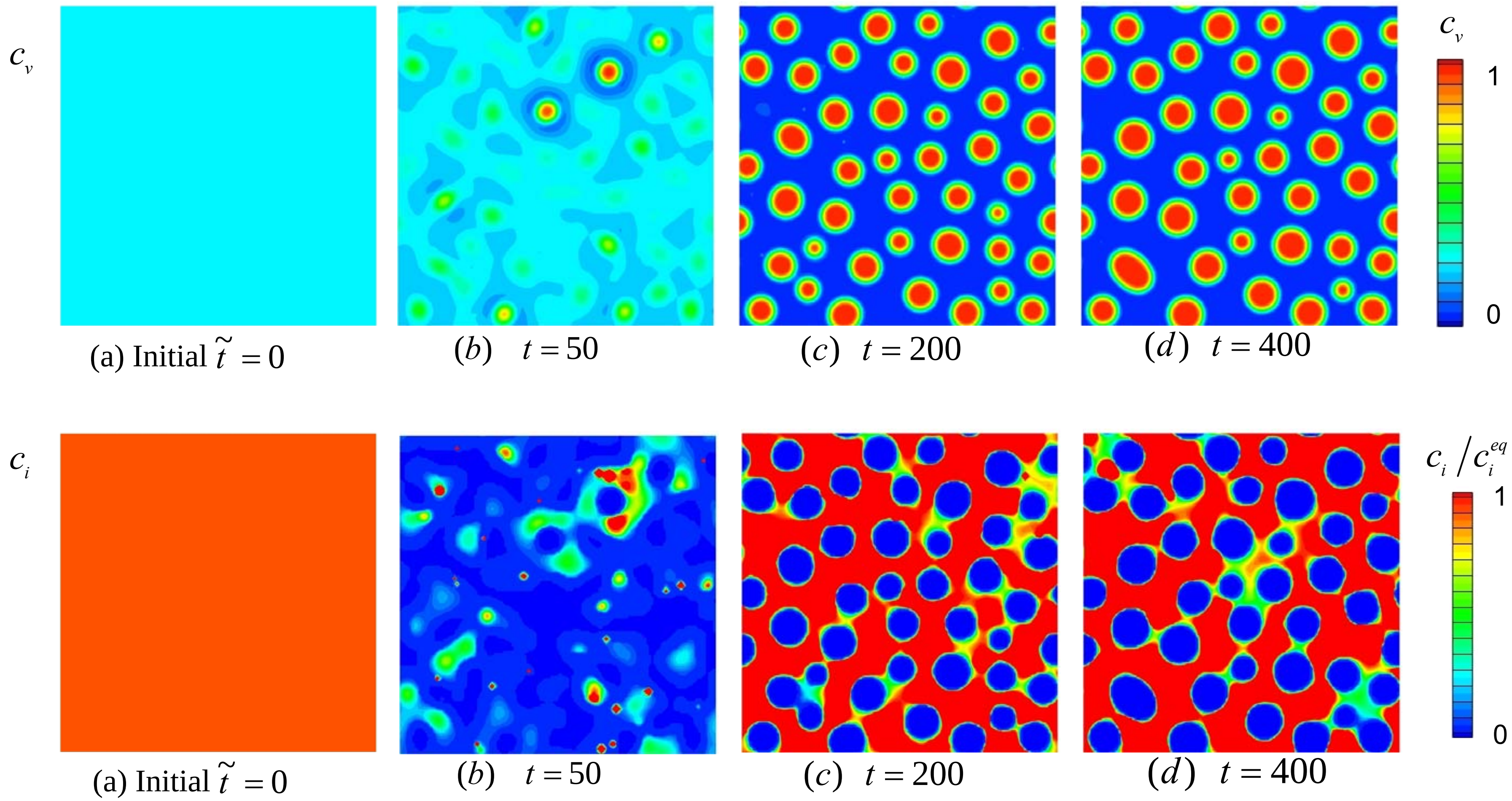
Effect of thermal fluctuations on void growth under irradiation



Vacancy and interstitial field evolution showing void growth in the presence of radiation effects and thermal fluctuations.

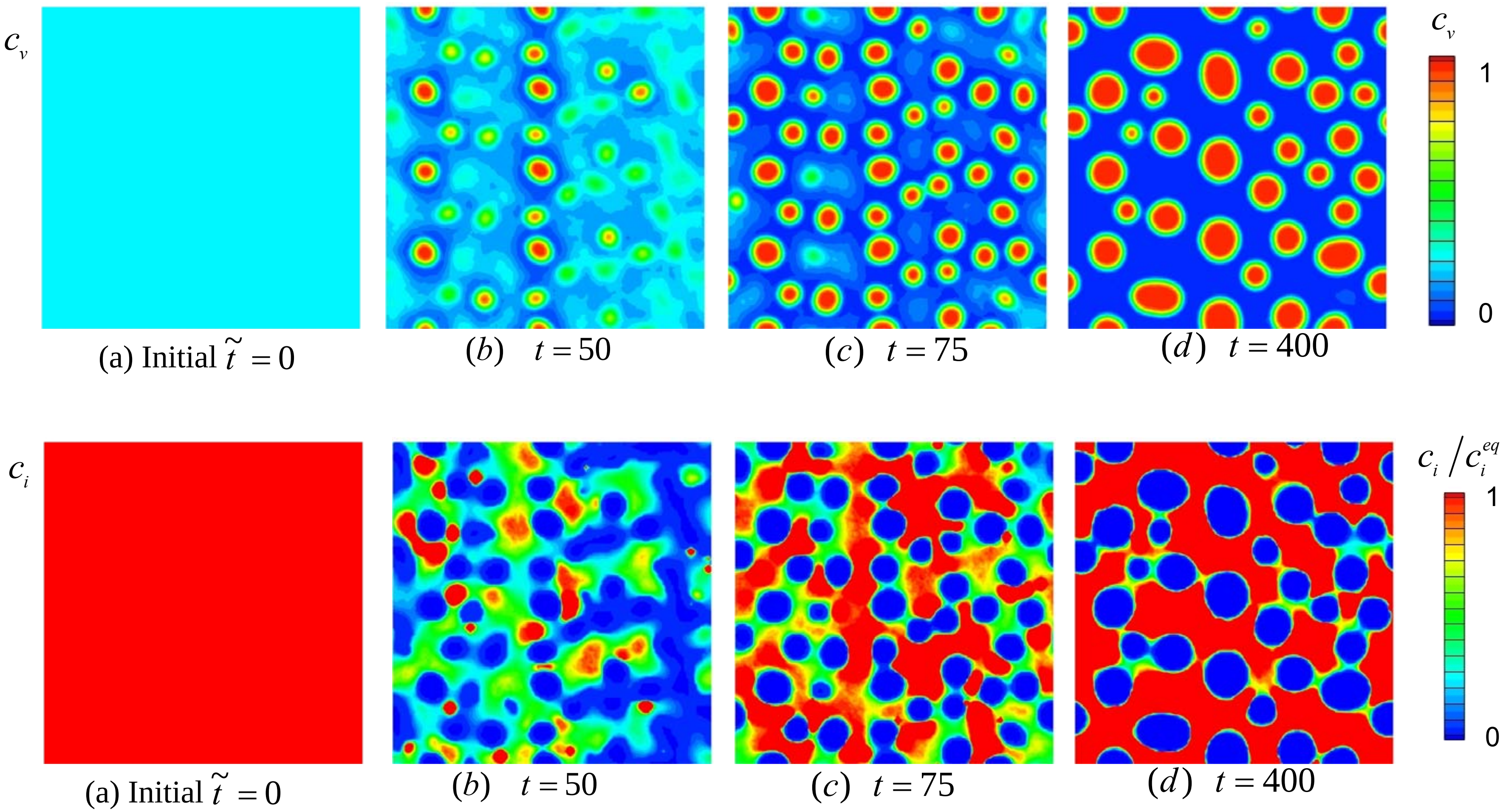
S_v=50, S_i = 1, P_v=0.25, P_i = 0.15 on 128 x 128 grid

Void nucleation and growth due to irradiation



Vacancy and interstitial field evolution showing void nucleation due to radiation effects. S_v=50, S_i = 1, P_v=0.25, P_i = 0.15 on 128x128 grid

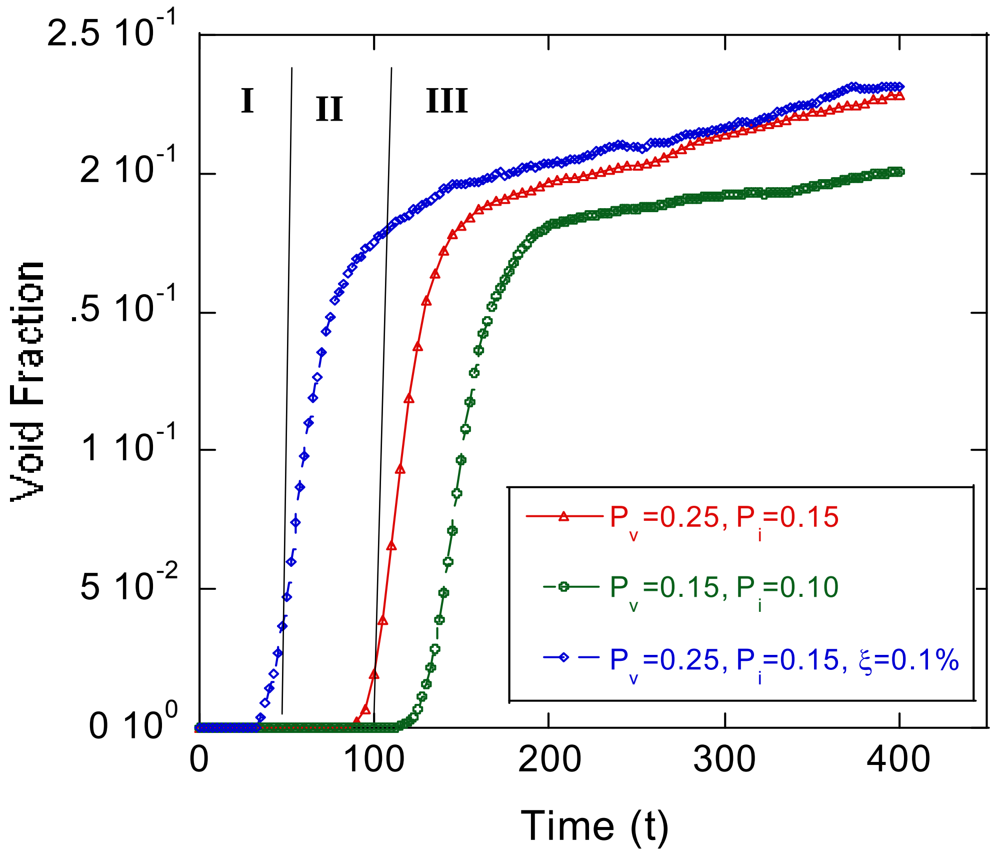
Effect of thermal fluctuations on void nucleation and growth



Vacancy and interstitial field evolution showing void nucleation radiation effects and thermal fluctuations.

S_v=50, S_i = 1, P_v=0.25, P_i = 0.15 on 128 x 128 grid

Analysis of Nucleation and Growth



Stage I : Incubation period

Stage II: Nucleation regime (Johnson-Mehl-Avrami Equation)

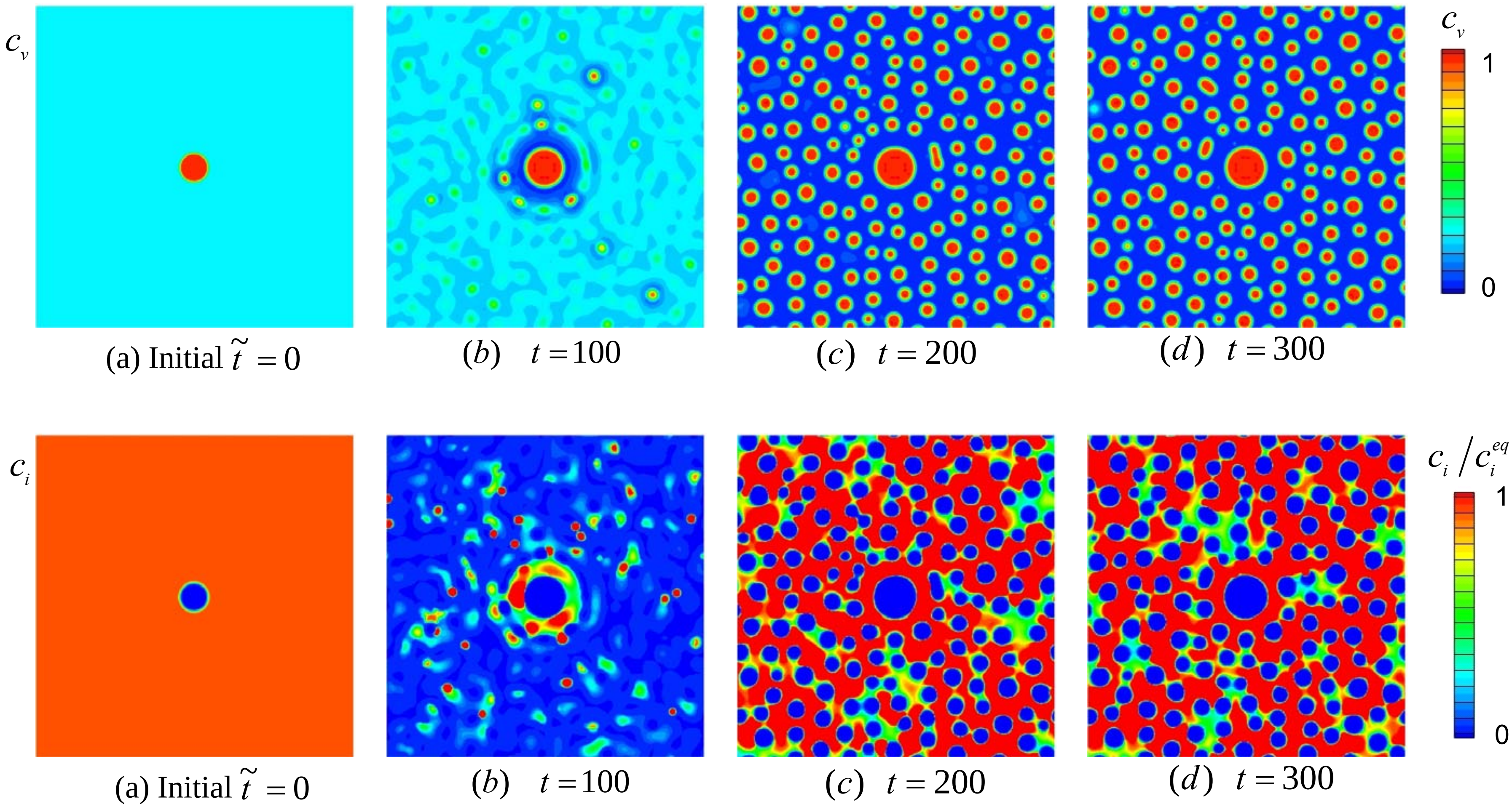
$$p = p^e\left(1-\exp\left(-kt^3\right)\right)$$

Stage III: Growth regime (Ostwald ripening)

$$p = p^o\left(1+t/\tau\right)^1$$

NOTE: The change in incubation time with decrease of cascade size and with thermal fluctuations

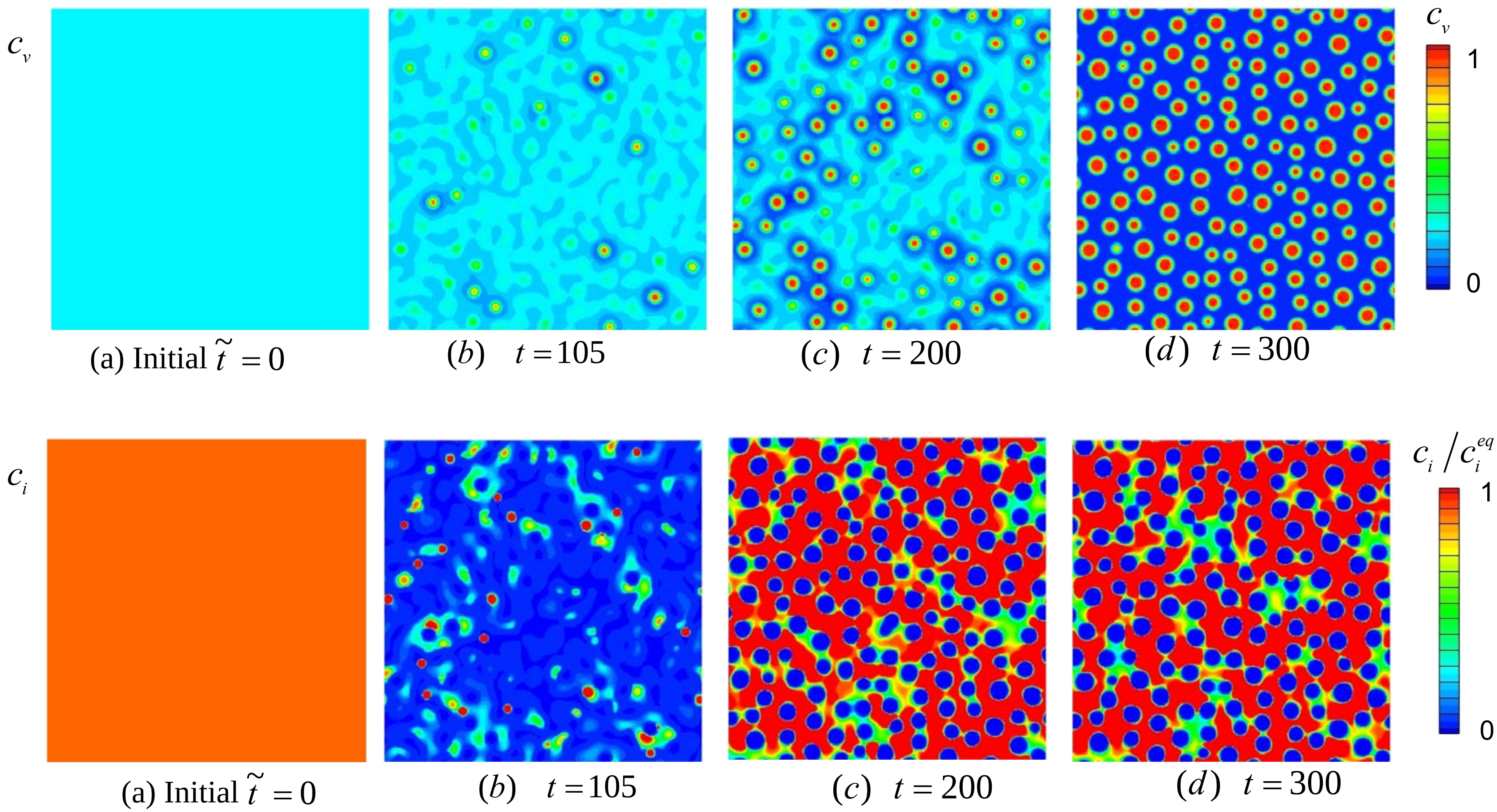
Void growth under irradiation



Vacancy and interstitial field evolution showing void growth in the presence of radiation effects.

S_v=50, S_i = 1, P_v=0.25, P_i = 0.15 (on 256 x 256 grid), r_{void} = 10

Void nucleation under irradiation



Vacancy and interstitial field evolution showing void nucleation presence of radiation effects.

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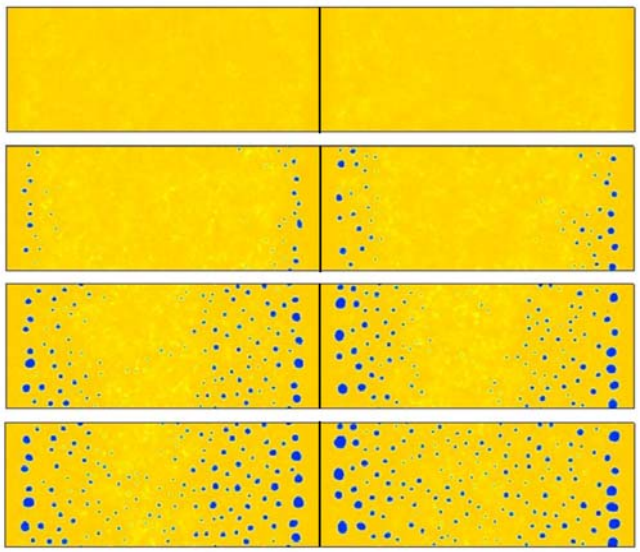
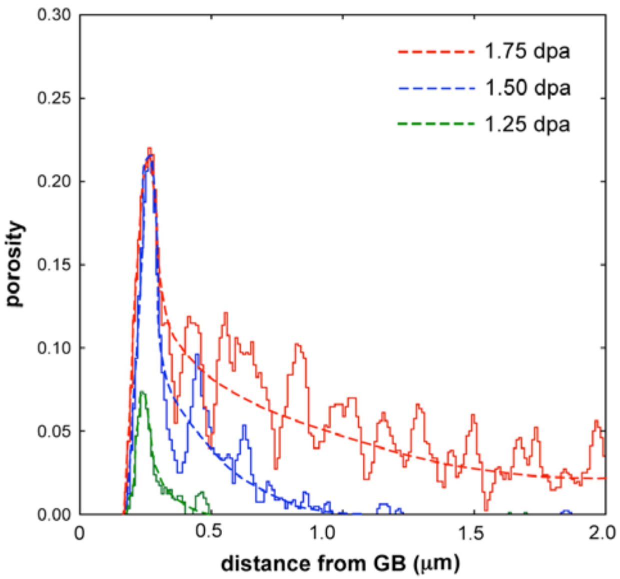
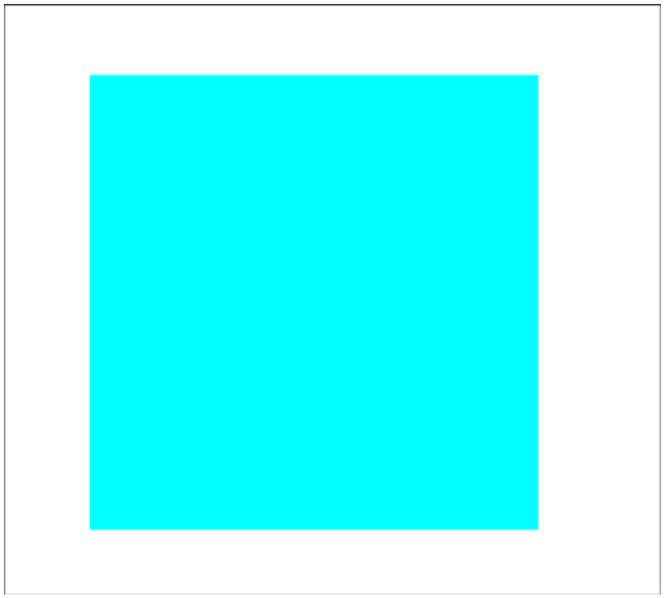
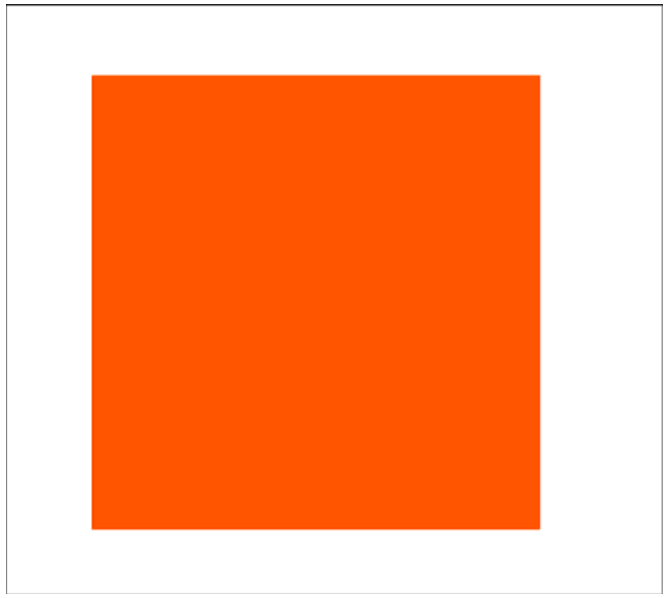
Role of grain boundaries





Nucleation and growth (movies)





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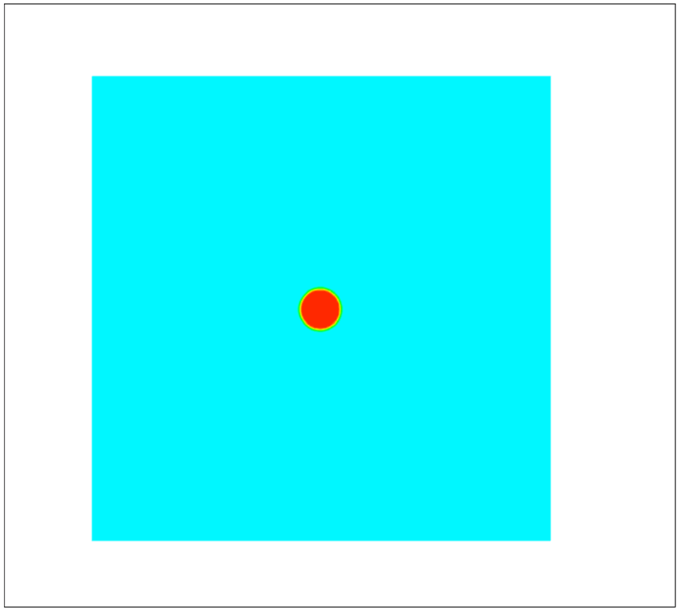
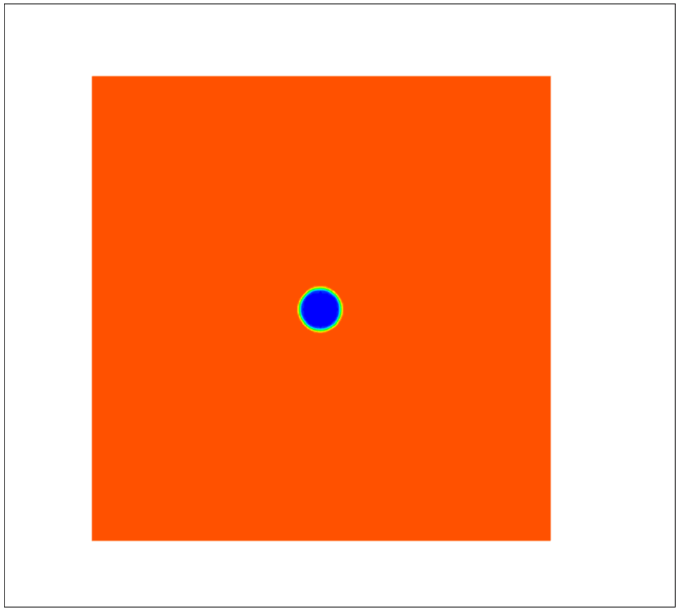
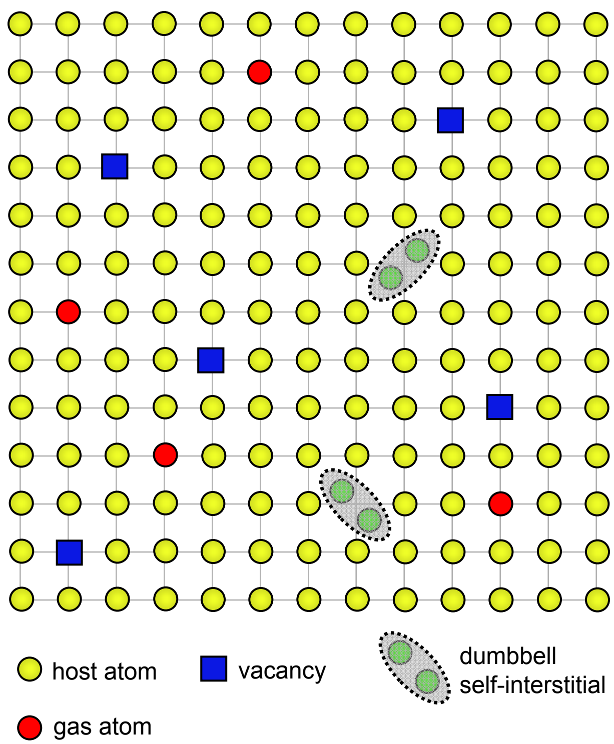




Gas effects and bubble formation



The model has been extended to include gas atoms and to model the nucleation and growth of gas bubbles. Preliminary results show good agreement with experimental observations

Summary

- A phase field model for void/bubble nucleation and growth
- Vacancies, interstitials, gas atoms represented
- Models seems to predict the defect, void and bubble dynamics under irradiation

In progress

- **Current model:**
  - **Thin interface analysis to fix parameters and apply to real materials**
  - **Model dislocation loop nucleation**
  - **Add stress effects and diffusion anisotropy (capture void lattices)**
  - **Anisotropic surface energy – directional dependence of gradient energy term**
- **Generalization to multi-component systems**