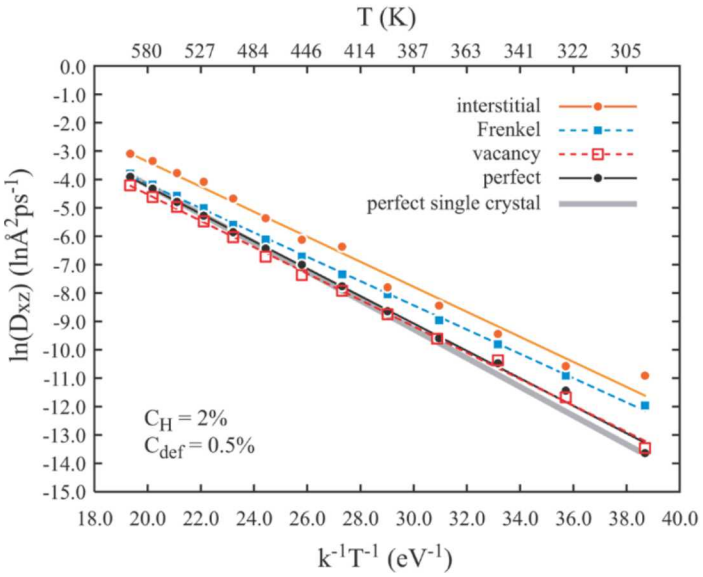
Molecular Dynamics Simulations of Radiation Effects on Hydrogen Isotope Diffusion in Stainless Steels

SAND2019-0533PE

Incorporating Statistics



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S&T Breakout, QMM

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Leidos Facility

2nd Floor Conference Room

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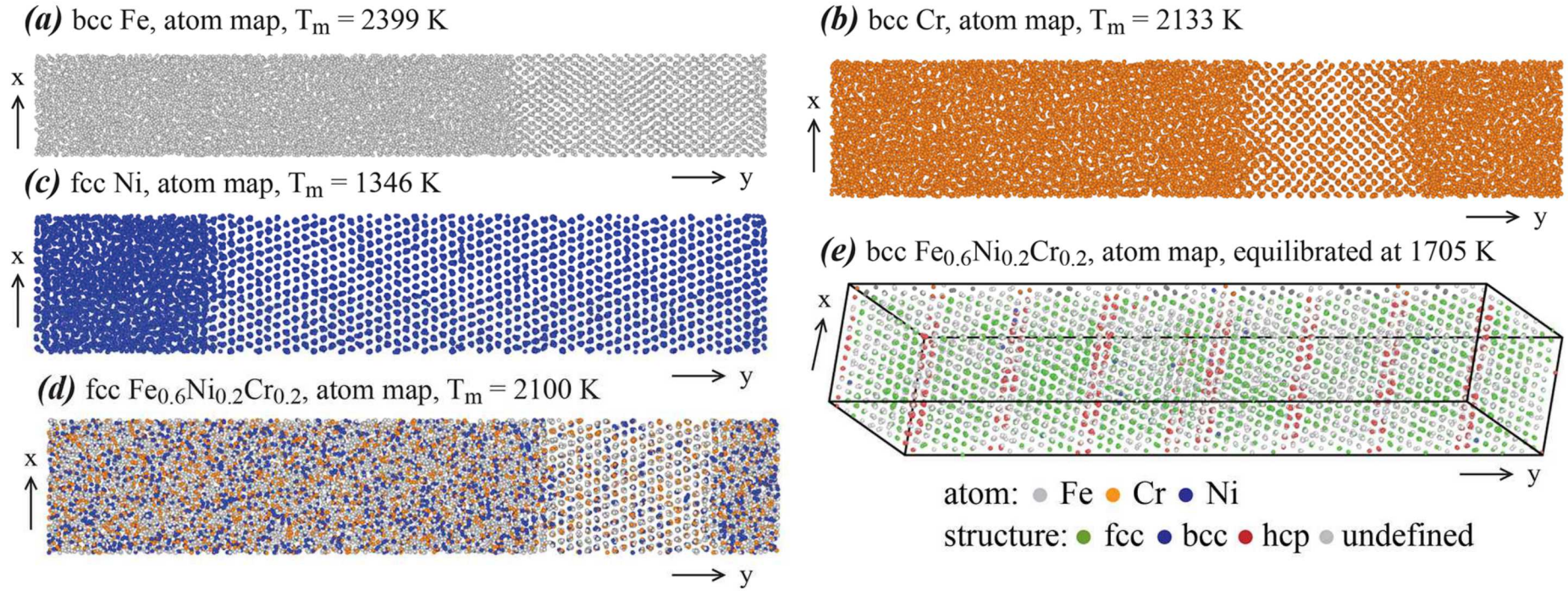
Motivation

- The TMIST-2 irradiation experiment at the Advanced Test Reactor at Idaho National Laboratory measured a tritium permeation enhancement in 316 stainless steel by a factor of ~2 to 5 relative to ex-reactor results.
- The Fe-Ni-Cr-H potential we recently developed enables molecular dynamics (MD) studies of hydrogen isotope diffusion in stainless steels (no other potentials enable this).

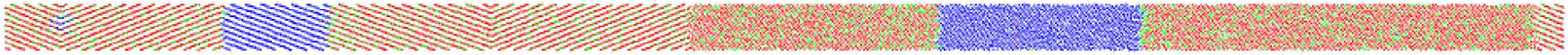
Important Notion:

Statistics of diffusion cannot be captured by DFT calculations. (MD) calculations of Arrhenius are required to understand this.

Fe-Ni-Cr-H potential enables MD simulations



Bonny et al's Fe-Ni-Cr potential (MSMSE 2013, 21, 085004) gives:

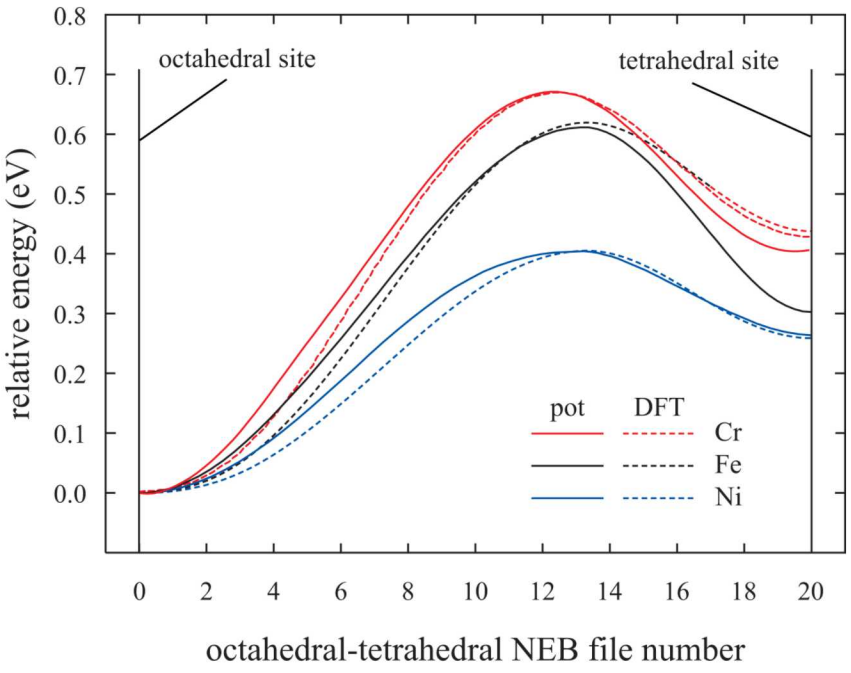


Fe-Ni-Cr-H potential gives the correct trends on H-M and H-H energies

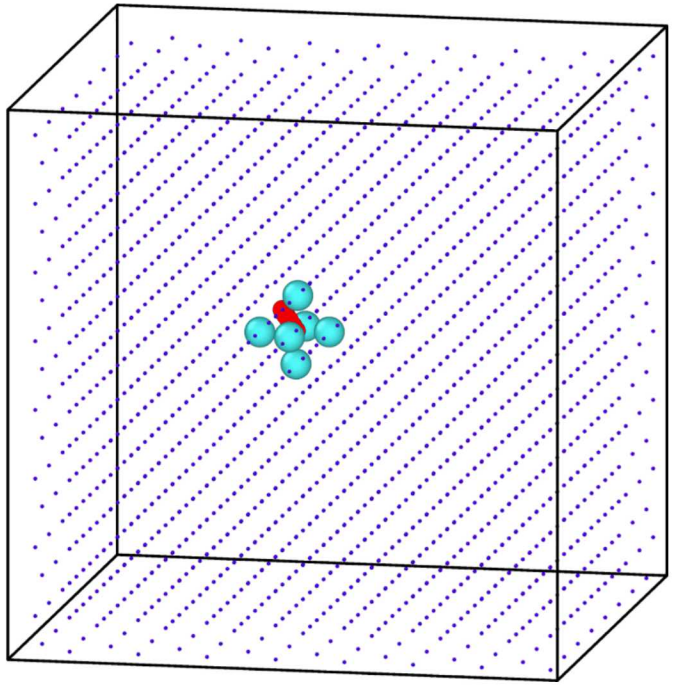
	Fe		Ni		Cr in Fe matrix	
	pot	DFT	pot	DFT	pot	DFT
$\Omega_{H,O}$	2.11	2.16	2.27	2.23	1.98	1.95
$E_{H,M}-E_{H,Fe}$	0.00	0.00	-0.12	-0.14	-0.29	-0.29
r_{H-H}	2.42	2.43	2.26	2.55	2.48	
ΔE_{H-H}	-0.015	-0.011	-0.074	-0.042	-0.208	
Q	0.61	0.62	0.40	0.40	0.67	0.67

Fe-Ni-Cr-H potential gives the correct H properties

Diffusion barrier Q_M under local M environment. Local Cr assumes fcc Fe matrix, otherwise local and matrix atoms are the same



Configuration for calculation Q_{Cr}: Small blue: Fe; big blue: Cr; red: H3



Planned work

- Perform MD simulations of hydrogen isotope diffusion in 316L stainless steels without defects and with manually created radiation defects (vacancies, interstitials, Frenkel pairs)
- Perform MD simulations of high energy impacts of 316 stainless steels to explore natural defects formed during irradiation
- Perform MD simulations of hydrogen isotope diffusion in 316L stainless steel samples that have gone through high energy impact simulations to explore effects of natural irradiated defects

Diffusion analysis

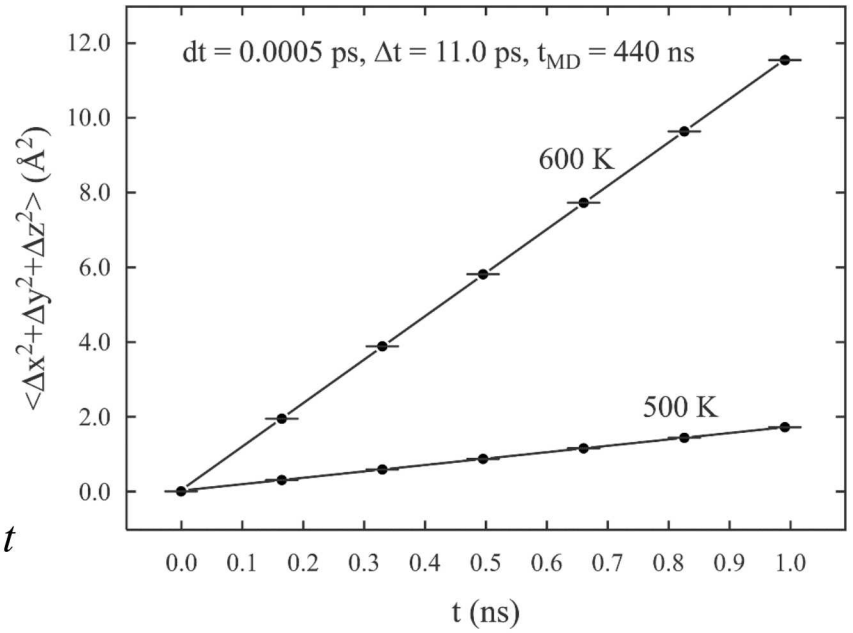
- The coordinates $\alpha_i(t)$ of N hydrogen atoms (i = 1, 2, …, N), are recorded on a time interval of Δt, i.e., at times of $t = j\Delta t$, j = 1, 2, …, m ($m = t_{MD}/\Delta t$), where Δt can be any multiple of the time step size dt used in the MD simulations.
- m+1-k measurements can be made for the displacement of a hydrogen atom i over a $k\Delta t$ period: $\Delta\alpha_{i,j}(k\Delta t) = \alpha_i(j\Delta t - \Delta t + k\Delta t) - \alpha_i(j\Delta t - \Delta t)$ where j = 1, 2, …, m+1-k.
- This allows us to calculate mean square displacement (MSD):

$$\left\langle \left[\Delta\alpha(k\Delta t)\right]^2 \right\rangle = \frac{\sum_{i=1}^{N}\sum_{j=1}^{m+1-k}\left[\Delta\alpha_{i,j}(k\Delta t)\right]^2}{N(m+1-k)}$$

- MSD can be fitted to diffusivities D:

$$\left\langle \left[\Delta\alpha(k\Delta t)\right]^2 \right\rangle = 2D_\alpha t$$
$$\left\langle \left[\Delta x(k\Delta t)\right]^2 \right\rangle + \left\langle \left[\Delta z(k\Delta t)\right]^2 \right\rangle = 4D_{xz} t$$
$$\left\langle \left[\Delta x(k\Delta t)\right]^2 \right\rangle + \left\langle \left[\Delta y(k\Delta t)\right]^2 \right\rangle + \left\langle \left[\Delta z(k\Delta t)\right]^2 \right\rangle = 6D_{xyz} t$$

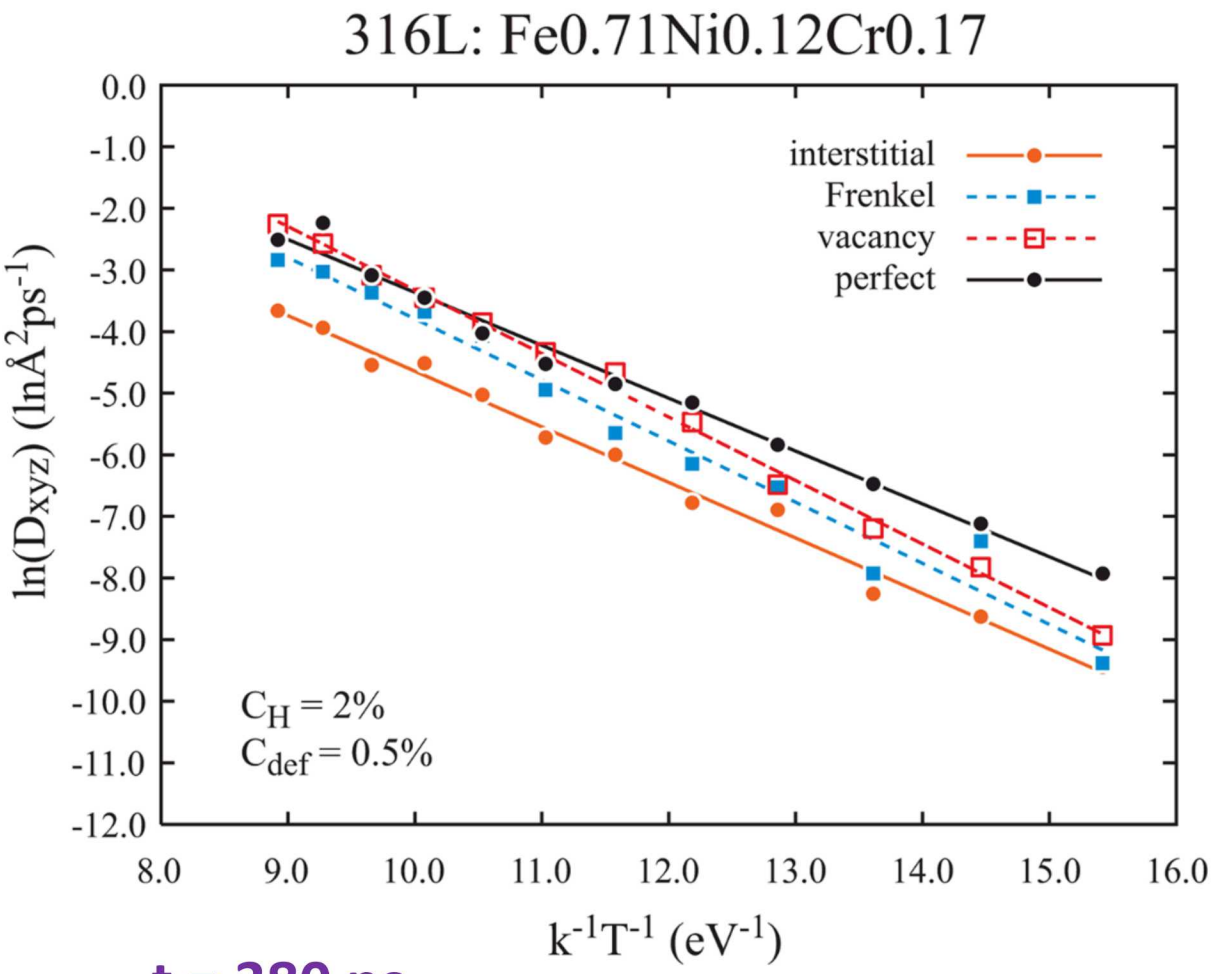
MSD convergence figure



Diffusion simulation conditions

- 316L composition (Fe0.71Ni0.12Cr0.17)
- Hydrogen concentration of C_H = 2%, defect concentration = 0.05%
- 22×25×21 $Å^3$ system size
- 380 ns simulation time (after the first 1 ns equilibration)
- 15 temperatures (600 K, 650 K, 700 K, …, 1300 K)

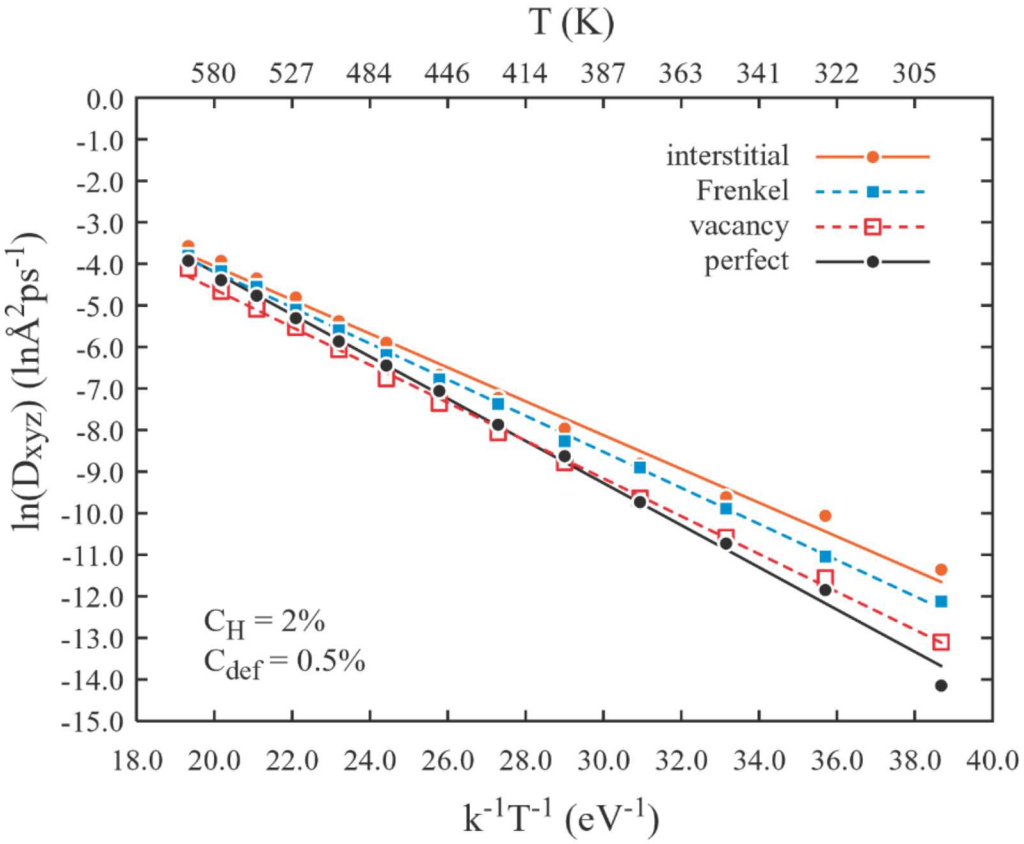
Arrhenius plots



t = 380 ns

- Defects reduce diffusivities
- Vacancy increases energy barrier, suggesting trapping

Arrhenius plots obtained previously for Ni

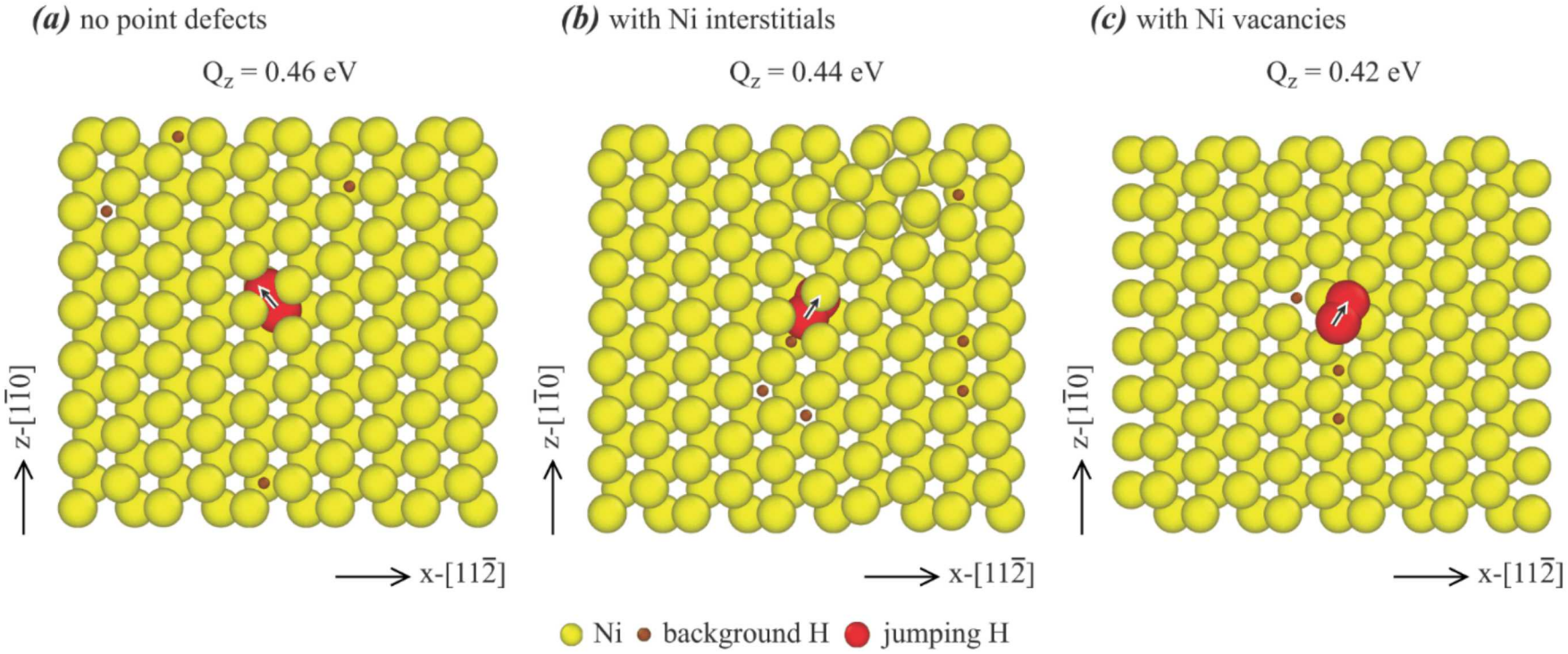


- Diffusivities are close with and without vacancies
- At the simulated interstitial concentration of 0.5%, interstitial increases diffusivities by 16.3 times at 300 K and 1.4 times at 600 K as compared with perfect crystals
- Activation energy of diffusion for perfect crystals is predicted to be 0.51 eV, as compared to the experimental value of 0.40 eV*

*L. Katz, M. Guinan, and R. J. Borg, Phys. Rev. B, 4, 330 (1971)

t = 440 ns

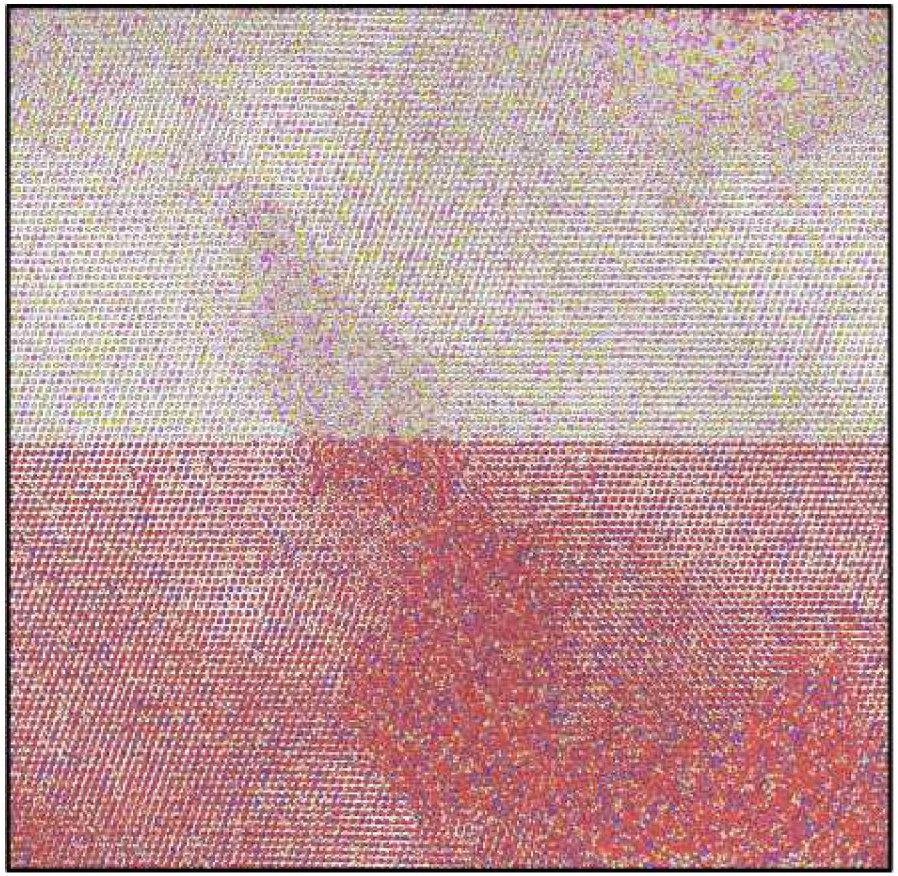
Atomic jumps in single crystalline Ni



Presence of nickel interstitials or vacancies can reduce the energy barrier of some hydrogen jump paths, but not too much

High energy impact simulations

- Initial problem of not forming cascade has been fixed
- Simulations of impacts at different energies are underway
- Results of these simulations will be used to further perform H diffusion simulations



316L ($Fe_{0.71}Ni_{0.12}Cr_{0.17}$, 2%H, 230×230×230 $Å^3$ size), 10 ps after impact at 0.3 MeV

Current status and future work

1. Simulations with pure Ni (literature potential) and 316L (our potential) revealed opposite effects of defects on H diffusion
2. To eliminate alloy effects, diffusion simulations with pure fcc Fe and pure fcc Ni using our potential are underway
3. Diffusion simulations with naturally irradiated samples will be conducted