# Fast, vacancy-free climb of dislocation loops in bcc metals

T D Swinburne[1], K Arakawa[2], H Mori[3], H Yasuda[3],
M Isshiki[4], K Mimura[4], M Uchikoshi[4] and S L Dudarev[1]

[1]Department of Theory and Simulation, CCFE
[2]Department of Materials Science, Shimane University
[3]Research Center for UHVEM, Osaka University
[4]Institute of Multidisciplinary Research for Advanced Materials, Tohoku University

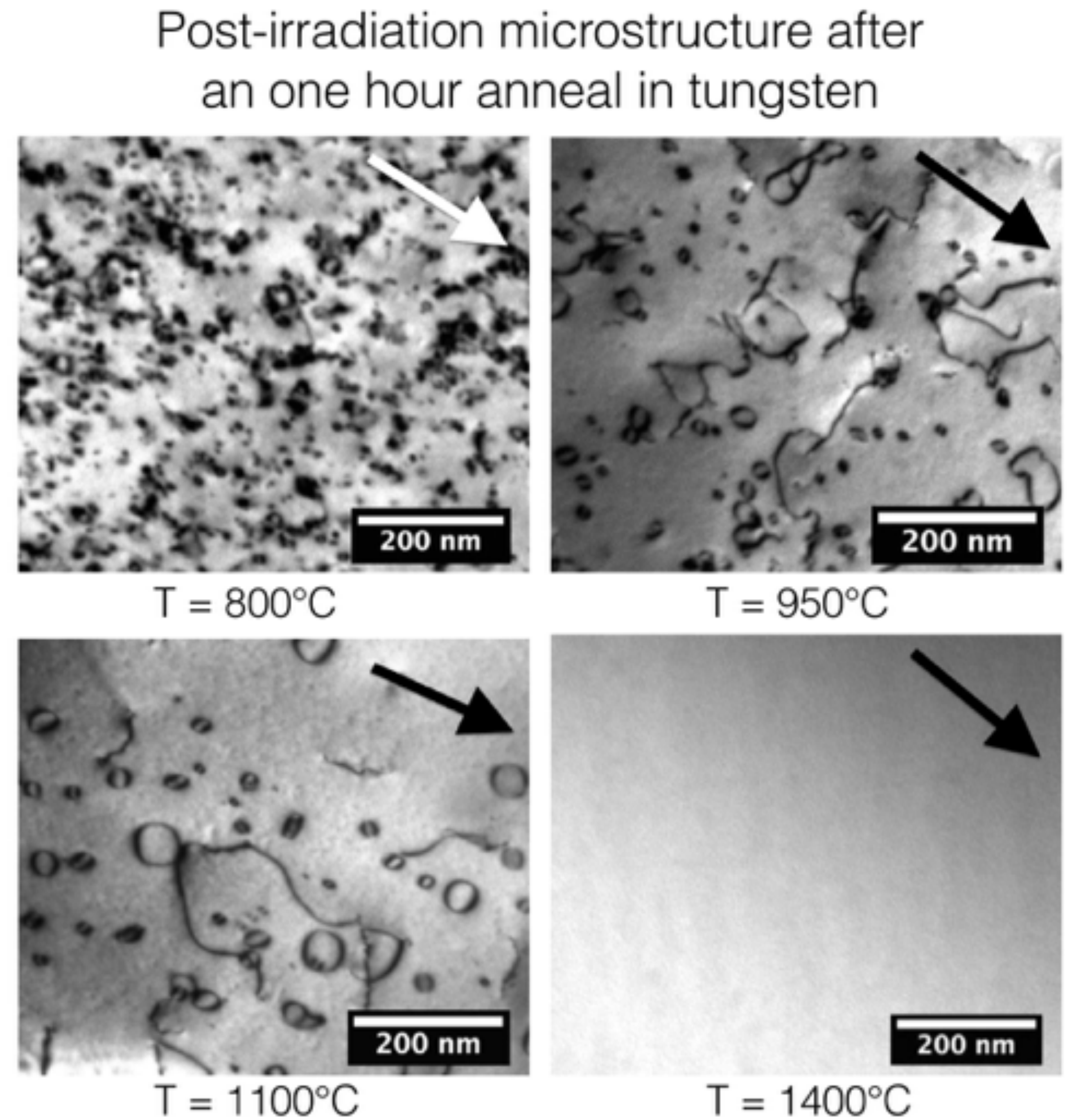
tomswinburne@gmail.com tiny.cc/tds110

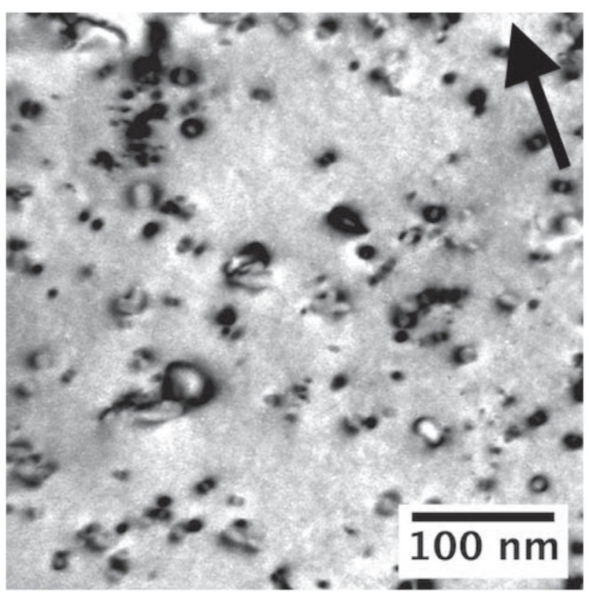
CCFE
CULHAM CENTRE FOR FUSION ENERGY

EUROfusion

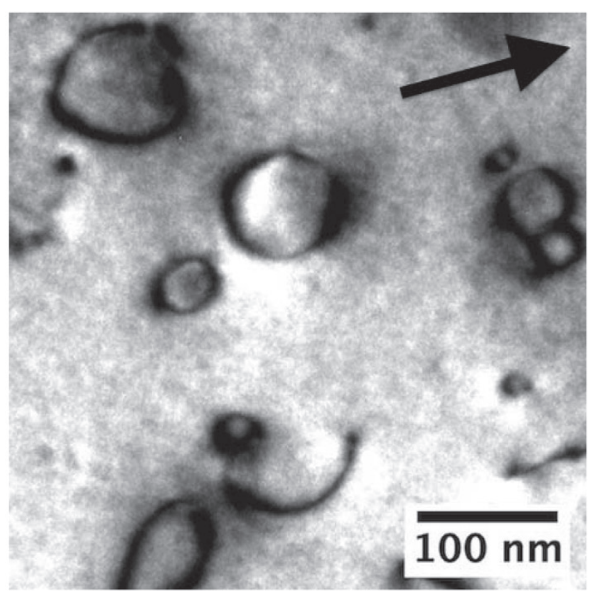
# Microstructure Evolution

- Post-irradiation annealing is known to be highly temperature dependent
- The dominant change in microstructure is the growth of prismatic dislocation loops




T=1097K, Δt=1 hour



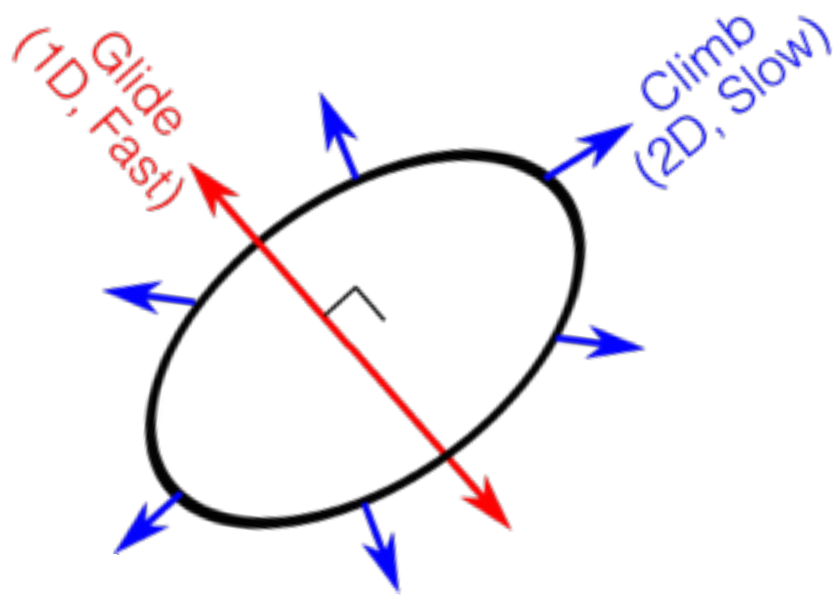

T=1097K, Δt=8 hours

F. Ferroni, X. Yi, K. Arakawa *et al.* Acta Met. 2015

# Climb

- Glide is rapid but often blocked by impurities / junctions etc

- Climb allows dislocations to leave their glide surface even when glide motion is blocked




- Climb typically requires **concurrent mass transport**, facilitated through biased diffusion of the vacancy atmosphere

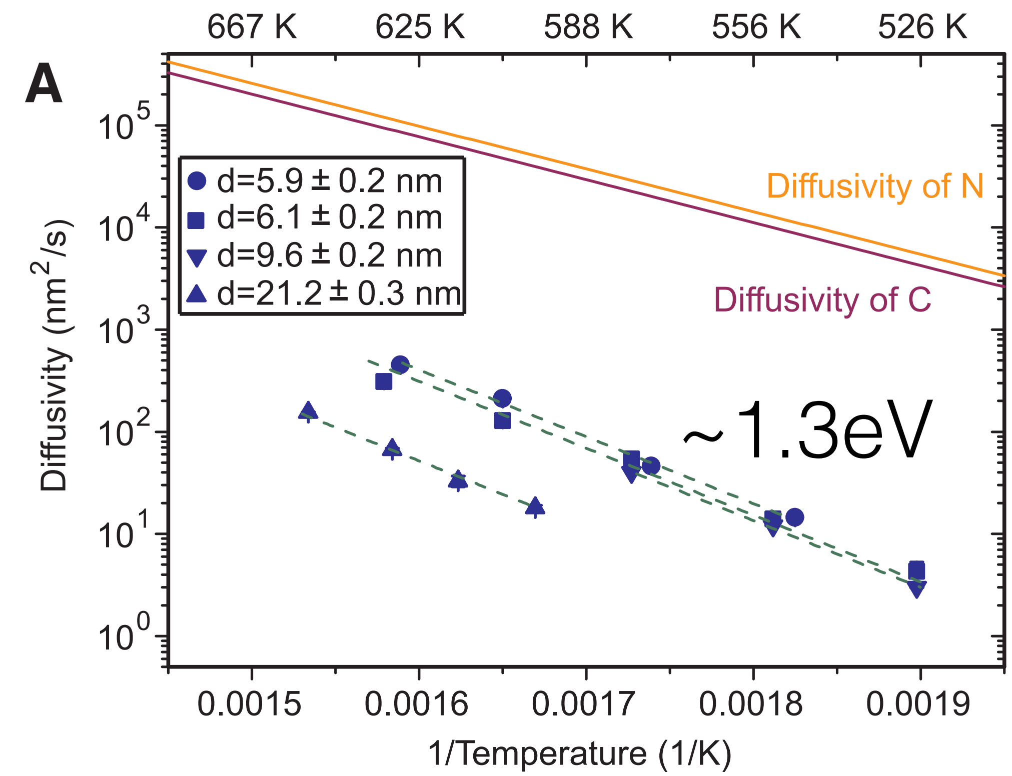



**Fig. 4.** Diffusivity of almost isolated ½⟨111⟩ loops.

Loop pinning in Fe by C / N
K. Arakawa *et al.* Science 2007

- Theory of **vacancy mediated climb (VMC)** gives

$$v_{\mathrm{VMC}} \propto c_{\mathrm{vacancies}} \times v_{\mathrm{vacancy}}$$
$$\propto e^{-\beta(E_{\mathrm{formation}}+E_{\mathrm{migration}})}$$

# Climb in post irradiation annealing

- Due to the restricted dimensionality of glide, climb has long been known to play an crucial role in PI annealing

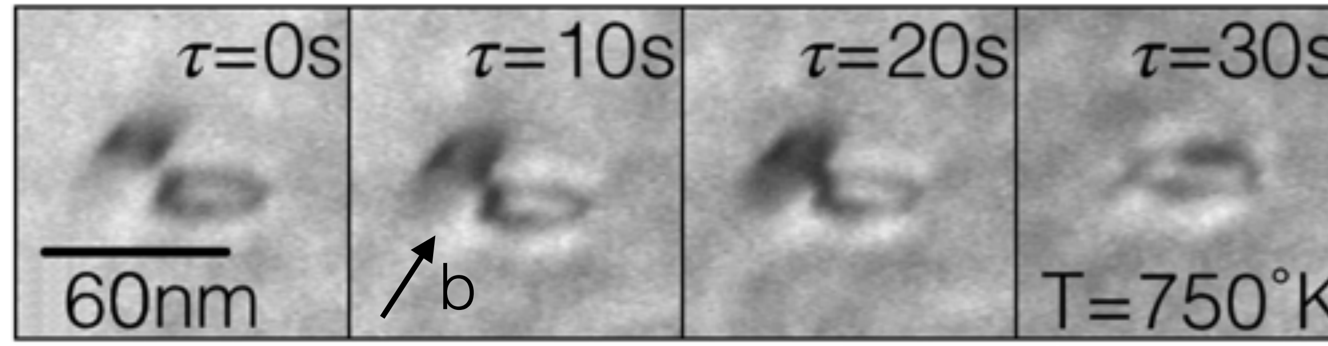
- This has been confirmed through **direct observation** under the TEM

- However, in many experiments climb motion is **up to x$10^6$ faster** than the predictions of climb theory

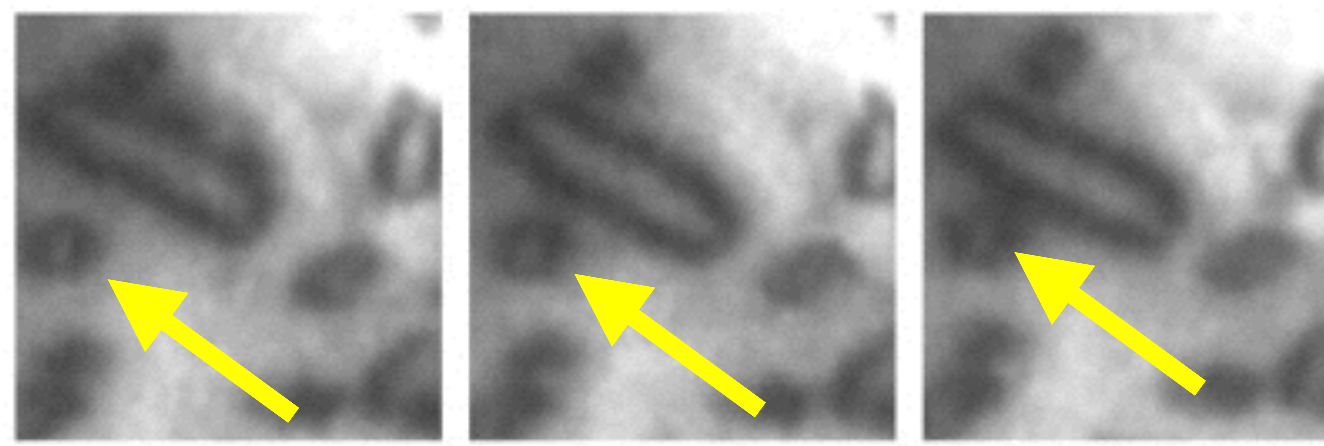
$$v_{\mathrm{VMC}} \propto e^{-\beta(E_{\mathrm{formation}}+E_{\mathrm{migration}}}$$

- In addition, the dislocation loops are actually executing non-glide motion, incompatible with climb theory.

1/2<111> loops in Fe à 750K (this work)




1/2<111> loops in W à 1273K
(Ferroni et al. Acta. Met. 2015)



(k) t=35 s (l) t=36 s (m) t=42 s

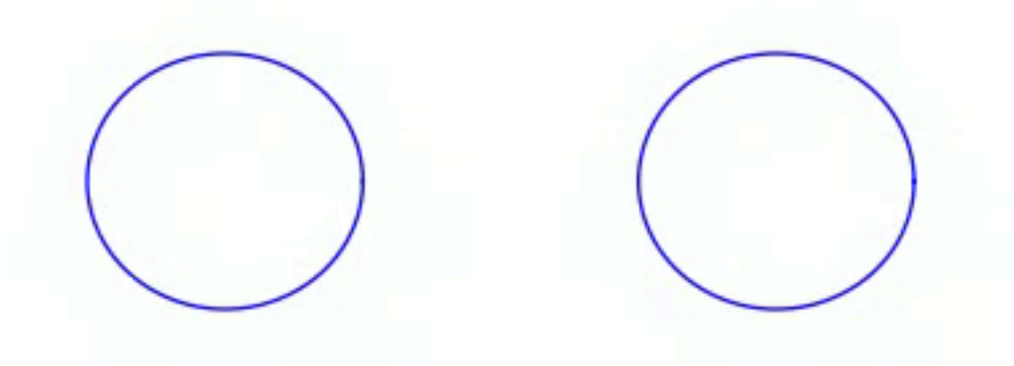
segment separations. For example, at 1200 °C two segments with 1 nm separation would coalesce solely by vacancy-mediated climb in ∼1 h, making this hypothesis very unlikely based on the experimental observations reported here.

F. Ferroni, X. Yi, K. Arakawa *et al.* Acta Met. 2015

# Climb in post irradiation annealing

- Vacancy mediated climb (VMC) is driven by biased diffusion of the surrounding vacancy atmosphere

- This leads to **non-local** coarsening of dislocation loops



VMC Mechanism

TEM Observations

# Climb in post irradiation annealing

| Feature | Vacancy Mediated Climb (VMC) | Experimental Observations |
|---|---|---|
| Loop growth mechanism | Growth by the evaporation of small loops due to non-local vacancy flux balance | $\neq$ Direct coalescence by non-glide motion, with no change in loop area |
| Rate of climb motion | $\sim 10^{-7}$Å$s^{-1}$ at $\sim 0.3T_m$ | $\ll$ $\sim 10^{0\text{-}1}$Å$s^{-1}$ at $\sim 0.3T_m$ |

- Vacancy supersaturation (due to e.g. irradiation) can in principle increase the VMC rate, but this cannot account for the mechanism of loop growth

- VMC cannot explain many experimental observations at these temperatures (it is still very important at higher temperatures!)

# Climb without vacancies

- This has been recognised since the 1960s, with an alternative mechanism, **self climb**, proposed to allow anomalously fast climb velocities

**The Growth of Prismatic Dislocation Loops during Annealing**

By C. A. Johnson‡
Materials Research Laboratory, University of California,
Berkeley, California

[Received June 20, 1960]

C. A. Johnson Phil. Mag. 1960

**The Coalescence of Dislocation Loops by Self Climb**

By J. A. Turnbull
Central Electricity Generating Board, Berkeley Nuclear Laboratories,
Berkeley, Gloucestershire GL13 9PB

[Received 15 September 1969]

J. A. Turnbull Phil. Mag. 1970

**Neutron Irradiation Damage in Molybdenum**
**Part V. Mechanisms of Vacancy and Interstitial Loop Growth during Post-irradiation Annealing**

By B. L. Eyre and D. M. Maher†
Metallurgy Division, Atomic Energy Research Establishment, Harwell, Berks.

[Received 6 April 1971]

Eyre and Maher Phil. Mag. 1971

**Self-climb of Dislocation Loops in Magnesium Oxide**

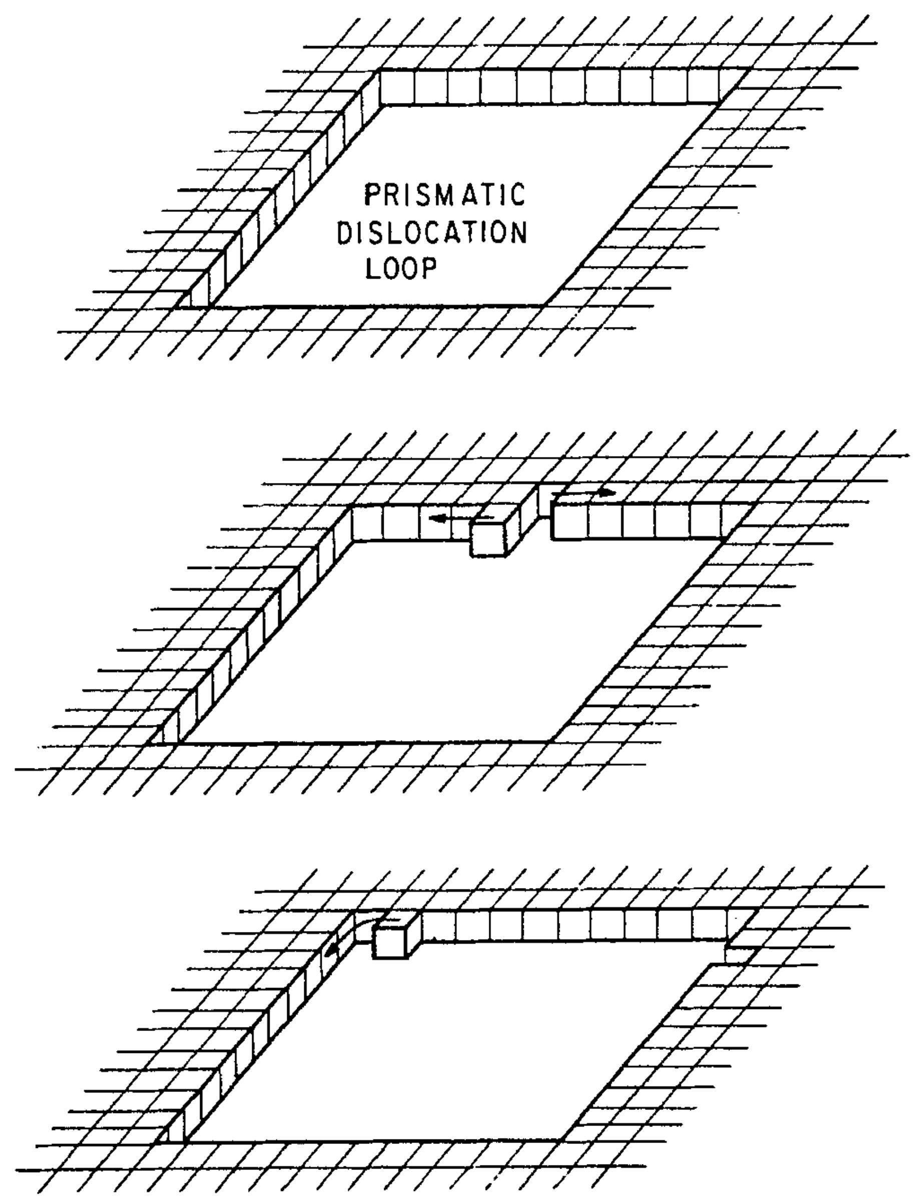
By J. Narayan and J. Washburn
Inorganic Materials Research Division, Lawrence Berkeley Laboratory,
Department of Materials Science and Engineering, College of Engineering,
University of California, Berkeley, California 94720, U.S.A.

[Received 1 February 1972 and in final form 19 June 1972]

Narayan and Washburn Phil. Mag. 1972

# Climb without vacancies

- In self climb, non-glide motion is driven through shape fluctuations (i.e. pipe diffusion) around the loop perimeter, much like the transport of surface islands




Formation and migration of defects involved in self-climb.

C. A. Johnson Phil. Mag. 1960

# Climb without vacancies

- In self climb, non-glide motion is driven through shape fluctuations (i.e. pipe diffusion) around the loop perimeter, much like the transport of surface islands

- It is simple to derive a self climb mobility -

$$\bar{x} = \sum_i \frac{x_i}{N}, \quad D_{\mathrm{SC}} = \lim_{t\to\infty} \frac{\langle \bar{x}^2 \rangle}{2t} = \frac{N_P}{N^2} D_P$$

$$\frac{\mathrm{v}_{\mathrm{climb}}}{\mathrm{f}_{\mathrm{climb}}} \equiv M_{\mathrm{SC}} = \beta D_{\mathrm{SC}} = \frac{2\beta a^5}{\pi R^3} \nu_0 e^{-\beta E_{\mathrm{SC}}}$$

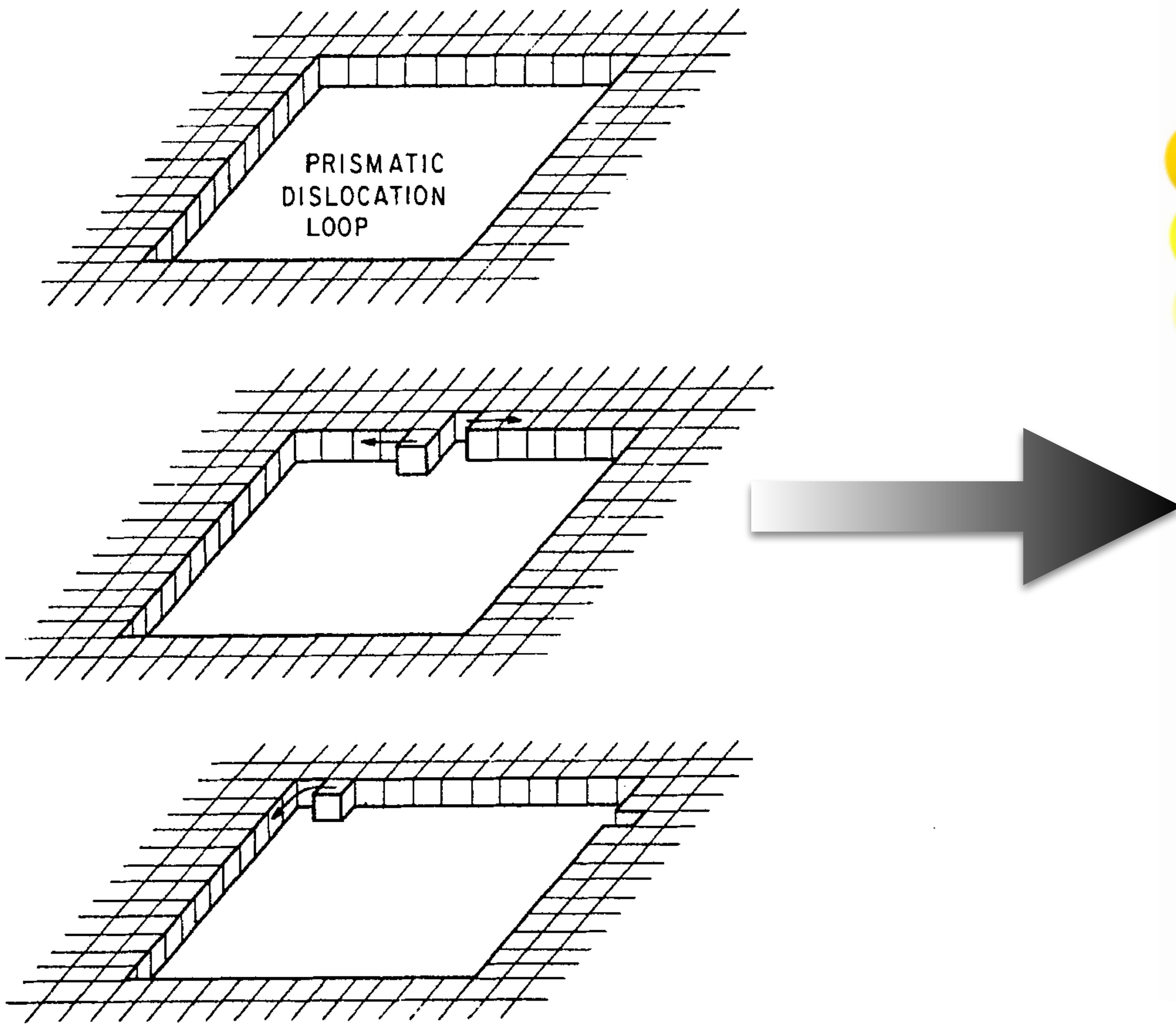
- But all previous studies could not calculate $E_{SC}$, the **critical, rate controlling parameter**

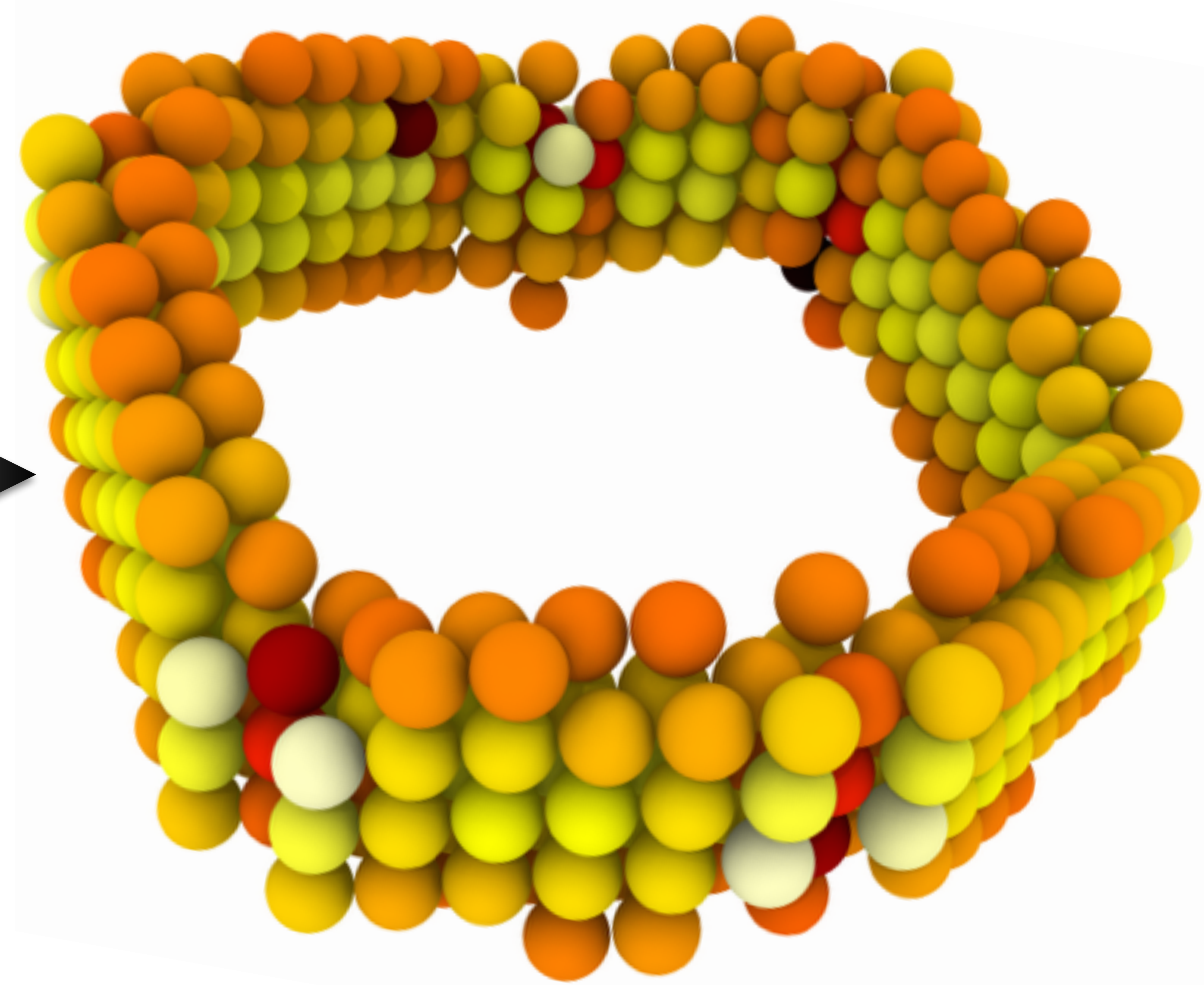
- Previous experiments (in Mo, MgO and Al) found results in the range

$$E_{\mathrm{SC}} \sim (0.4 - 0.7)(E_{\mathrm{formation}} + E_{\mathrm{migration}})$$

# Climb without vacancies

- To calculate a self climb activation energy, we looked at **structural fluctuations** of **irregular** 1/2<111> and <100> loops in Fe and W.



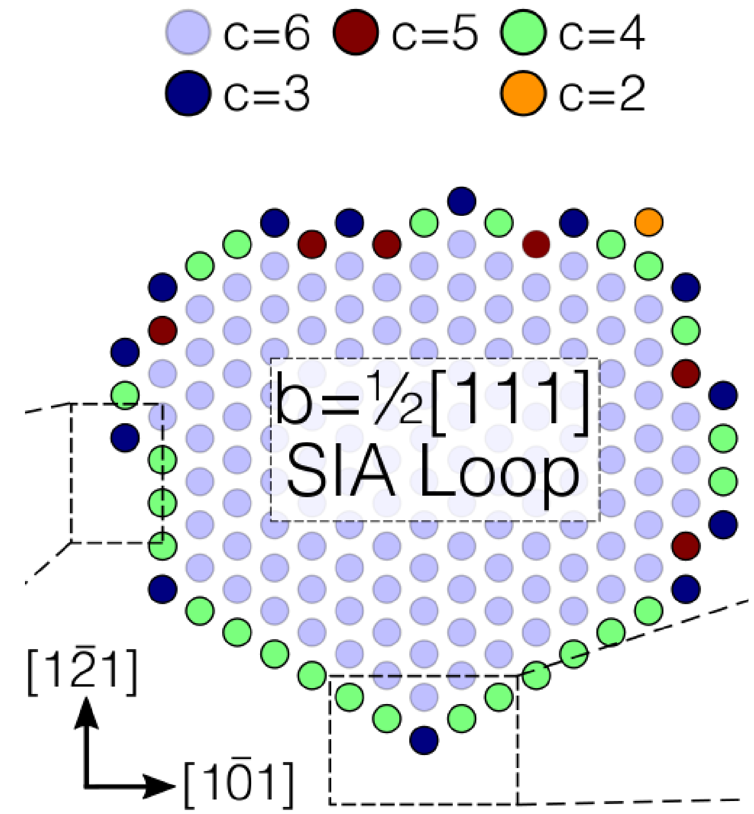



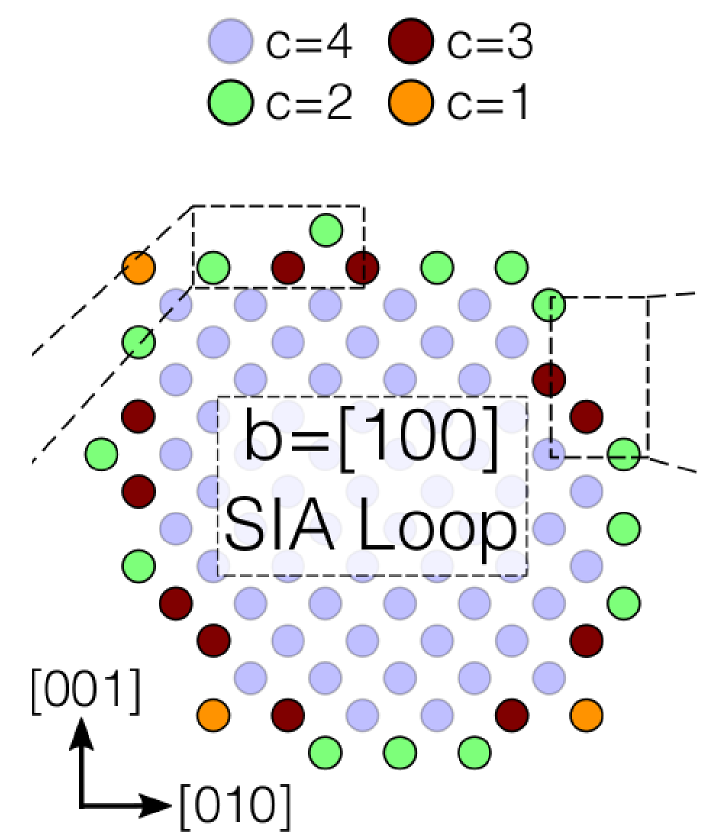
- Irregular loops are much more common than regular loops!

TDS, K Arakawa, *et al.*, *Scientific Reports* 2016

# Climb without vacancies

- Automated process written in LAMMPS/Python, using EAM potentials
  Fe: Gordon *et al.* Phil. Mag. 2011
  W: Marinica *et al.* JPCM 2013
- An irregular SIA palette (N~170) is constructed, inserted into a perfect lattice and relaxed
- Possible perimeter fluctuations of the initial palette are identified and then also formed in bulk
- NEB calculation to find migration pathway / barrier
- Identical calculations performed in a two loop supercell to gauge effect of nearby elastic fields (Answer: less than ~10% even with d = (2-3)a)



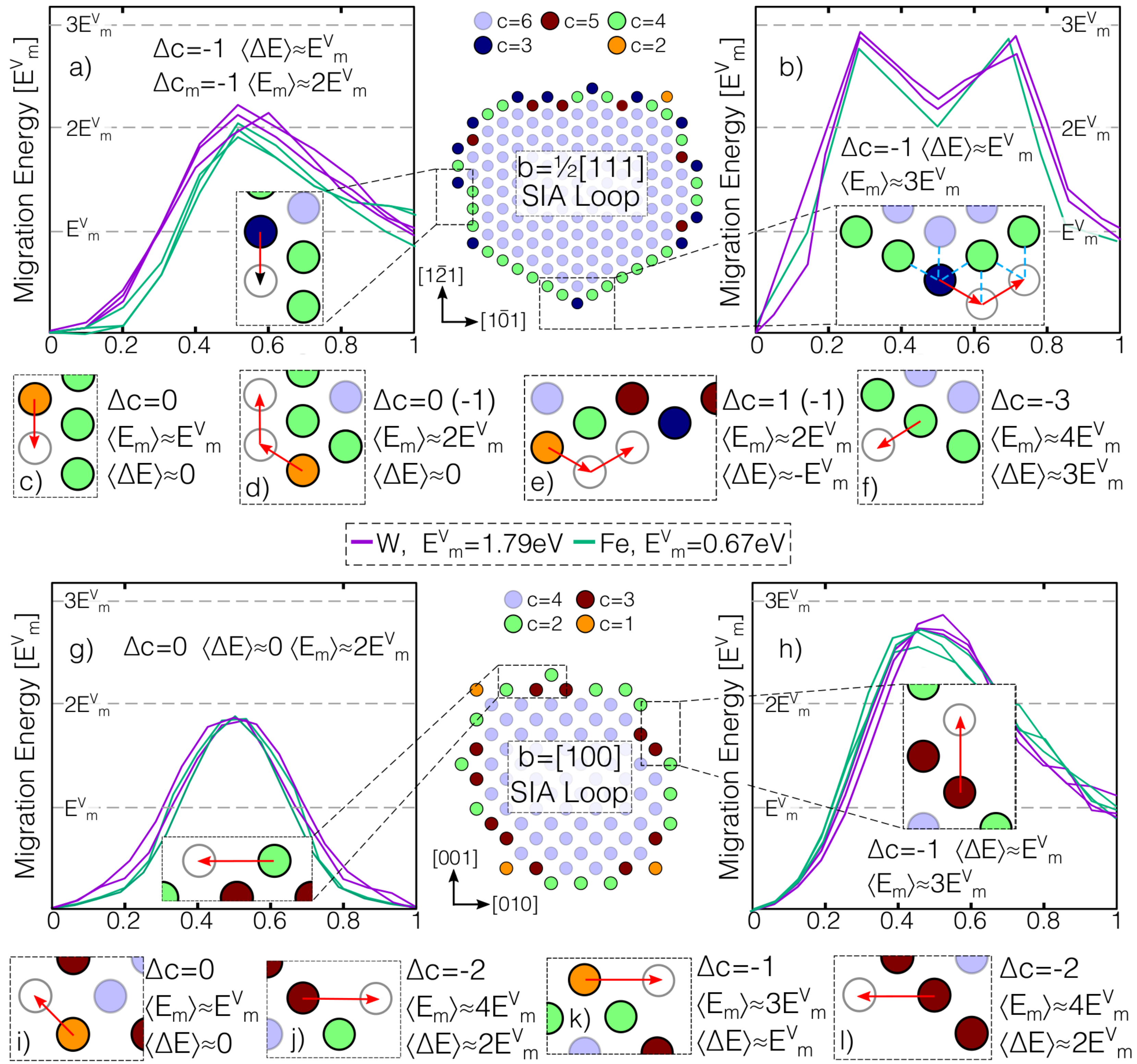




TDS, K Arakawa, *et al.*, *Scientific Reports* 2016

# Climb without vacancies

- All observed pathways could clearly be split into jumps along primitive lattice vectors 1/2⟨111⟩, ⟨111⟩, 1/2⟨111⟩

- The energy barriers in Fe,W could be normalised by the **vacancy** migration barrier




TDS, K Arakawa, *et al.*, *Scientific Reports* 2016

# Climb without vacancies

- From 200+ pathways, we found the simple average relation

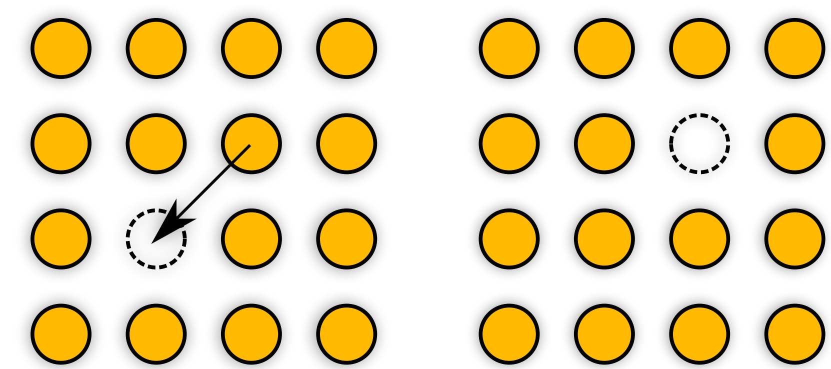
1/2<111>: $\Delta E \simeq \Delta c E^V_m, \quad E_m \simeq (\Delta c + 1) E^V_m$

<100>: $\Delta E \simeq \Delta c E^V_m, \quad E_m \simeq (\Delta c + 2) E^V_m$

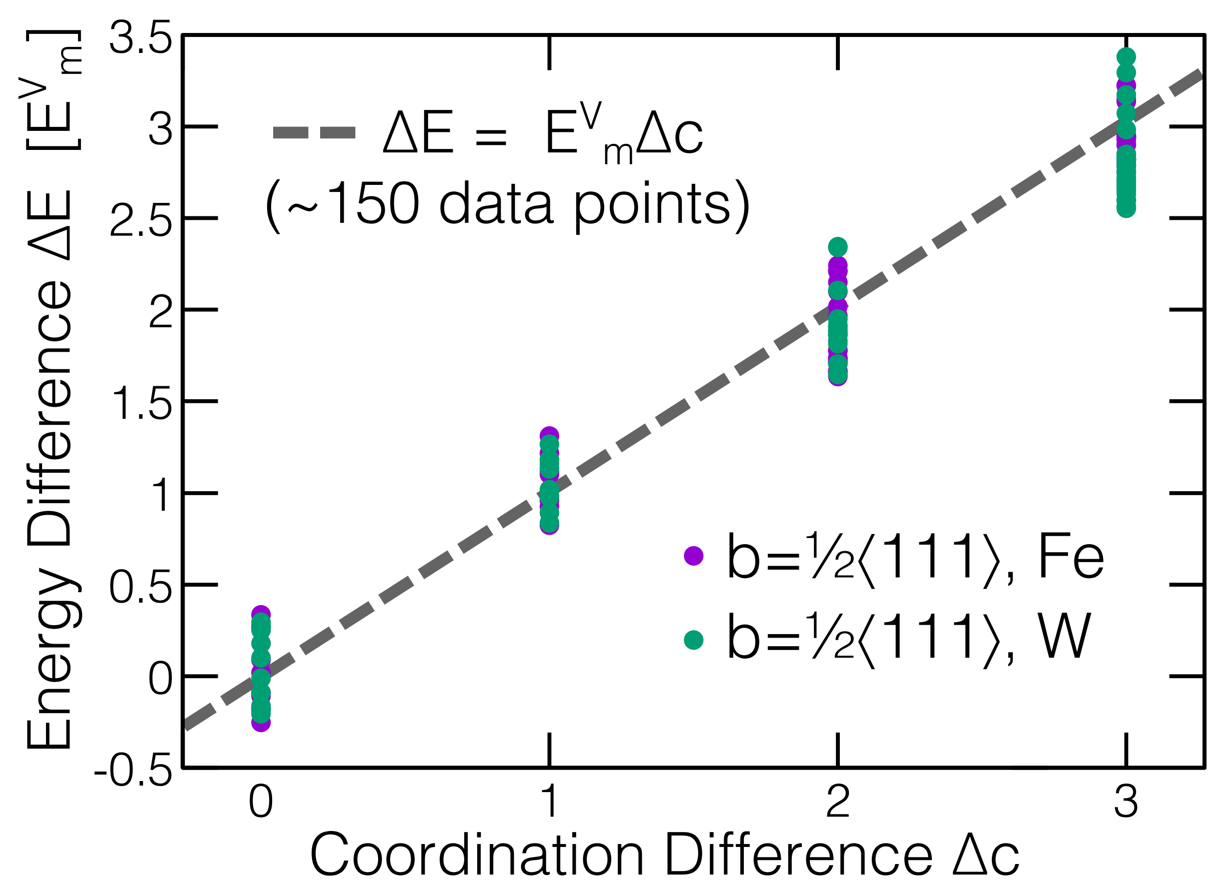
$E^V_m$ : **vacancy** migration barrier

$\Delta c$ : change in number of SIA neighbours

$$c_i = \sum_{j\neq i} \Theta\left(\frac{\sqrt{3}}{2}a - |\mathbf{x}_i - \mathbf{x}_j|\right)$$



Vacancy migration is the jump of a single atom




- The migration of a vacancy is the hopping of a single atom, `breaking` a nn `bond`
- For loop fluctuations we have a similar picture, with hops between SIA sites, with a similar **changes** in coordination

TDS, K Arakawa, *et al.*, *Scientific Reports* 2016

# Climb without vacancies

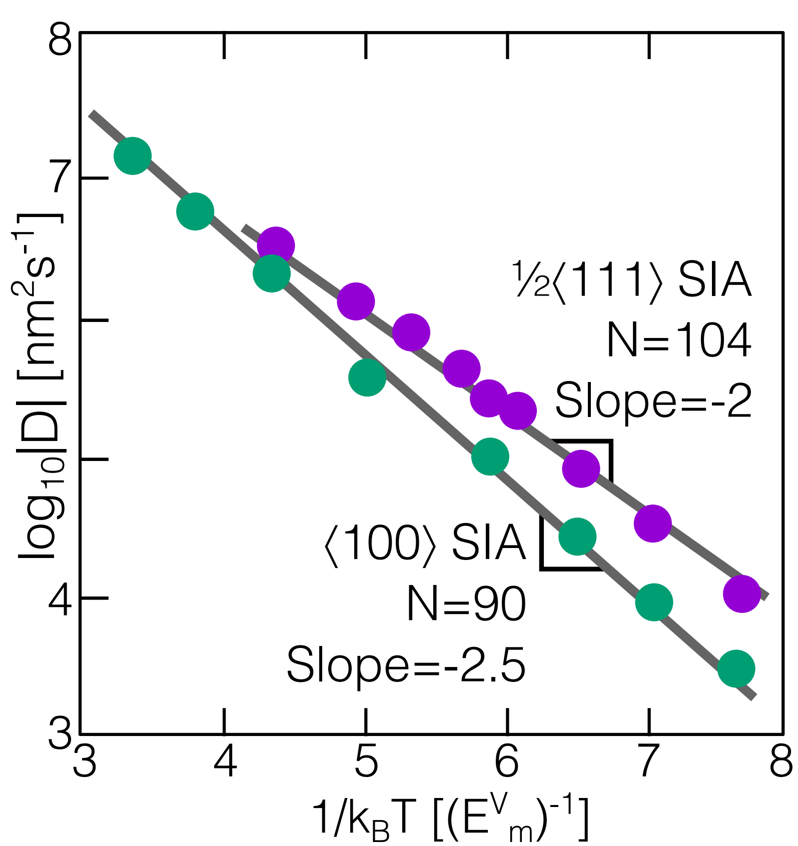
$$\Delta E = \Delta c E^V_m$$

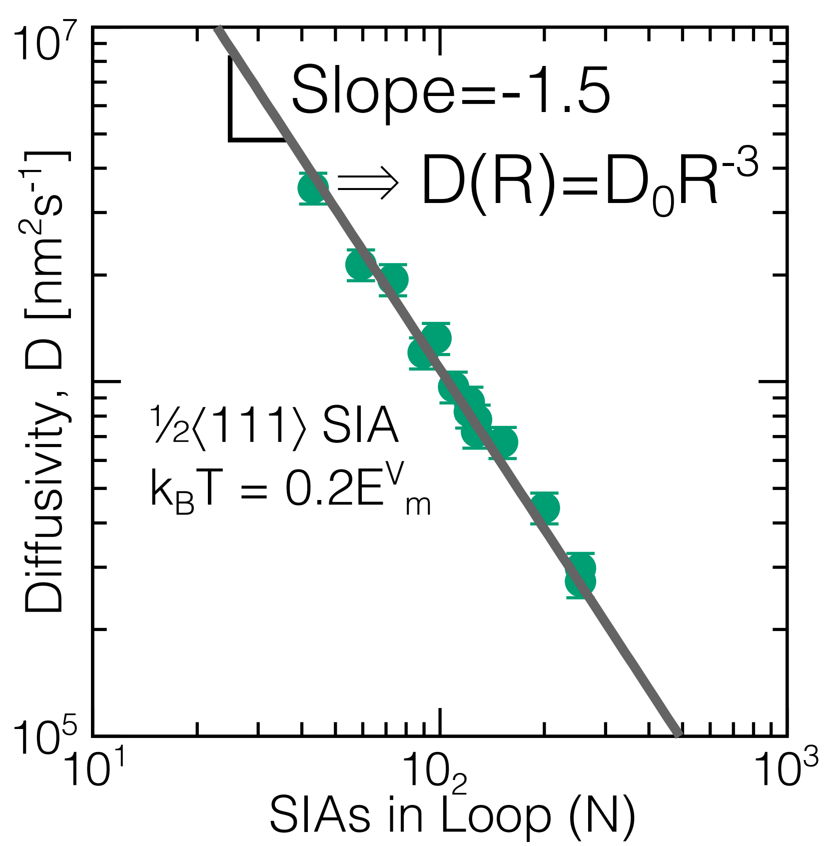
$$E^{\langle 111\rangle/2}_m = (\Delta c + 1)E^V_m$$
$$E^{\langle 100\rangle/2}_m = (\Delta c + 2)E^V_m$$

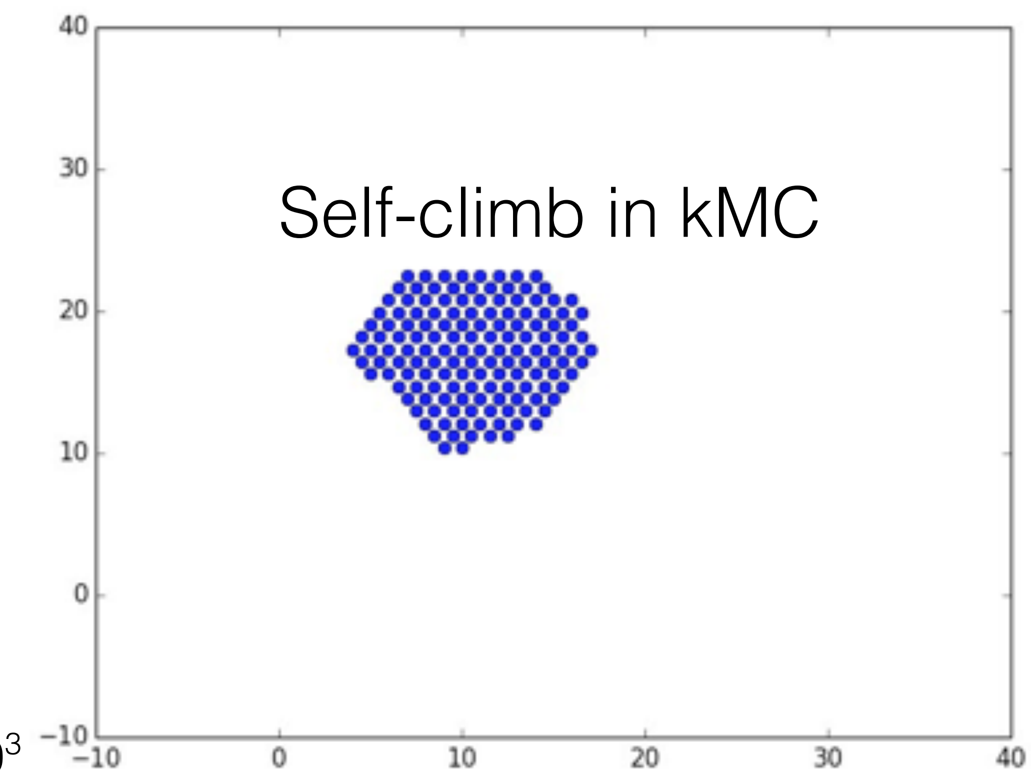
- Implementing this energy law in kMC we can calculate $D_{SC}$ and therefore $E_{SC}$, finding that

$$\mathrm{M}_{\mathrm{scl}} = \frac{2\beta\nu a^5}{\pi R^3} e^{-\beta E_{\mathrm{scl}}}$$

$$E^{\langle 100\rangle}_{\mathrm{scl}} = 2.5E^V_m,\ E^{1/2\langle 111\rangle}_{\mathrm{scl}} = 2.0E^V_m$$



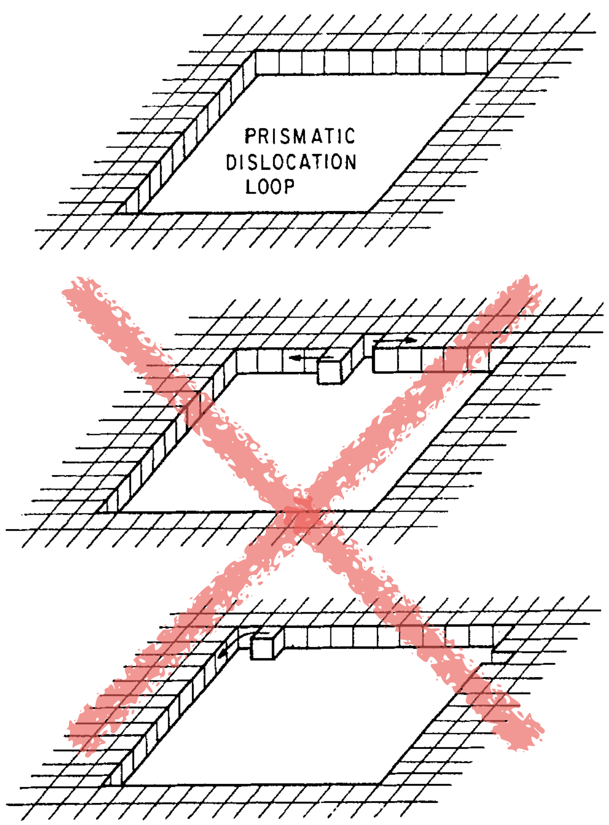



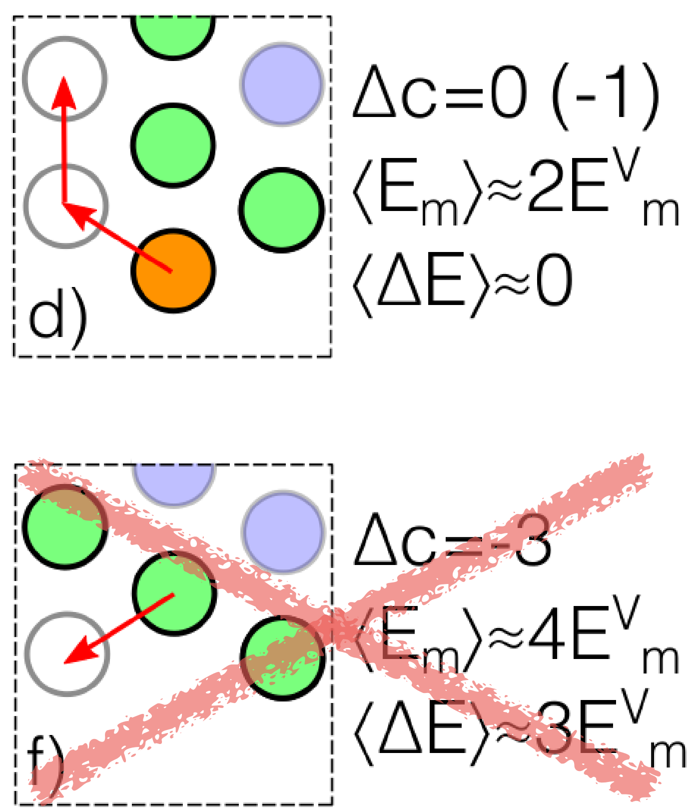

TDS, K Arakawa, *et al.*, *Scientific Reports* 2016

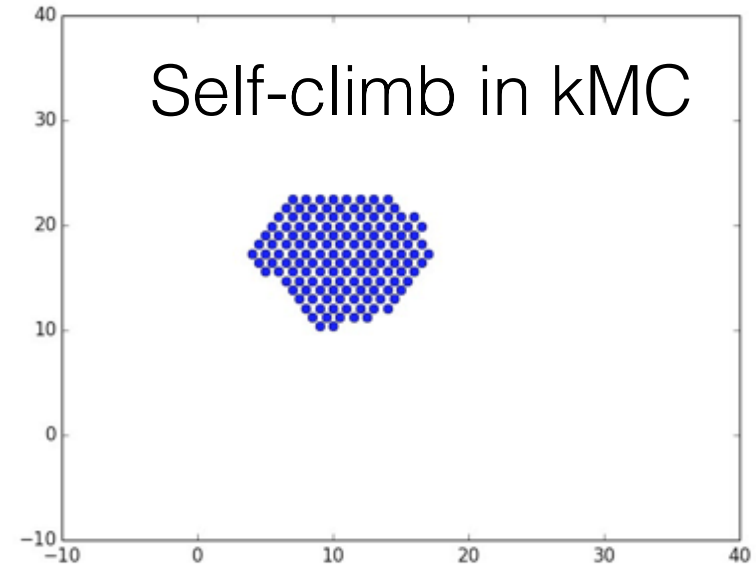
# Climb without vacancies

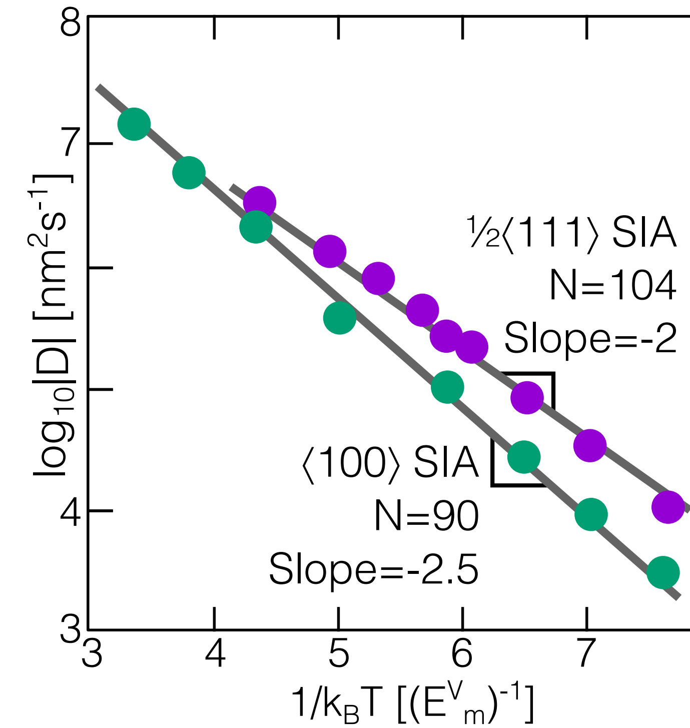
$$E_{\mathrm{scl}}^{\langle 100\rangle} = 2.5E_m^V,\; E_{\mathrm{scl}}^{1/2\langle 111\rangle} = 2.0E_m^V$$

- The activation energy corresponds to SIA migration around loop corners



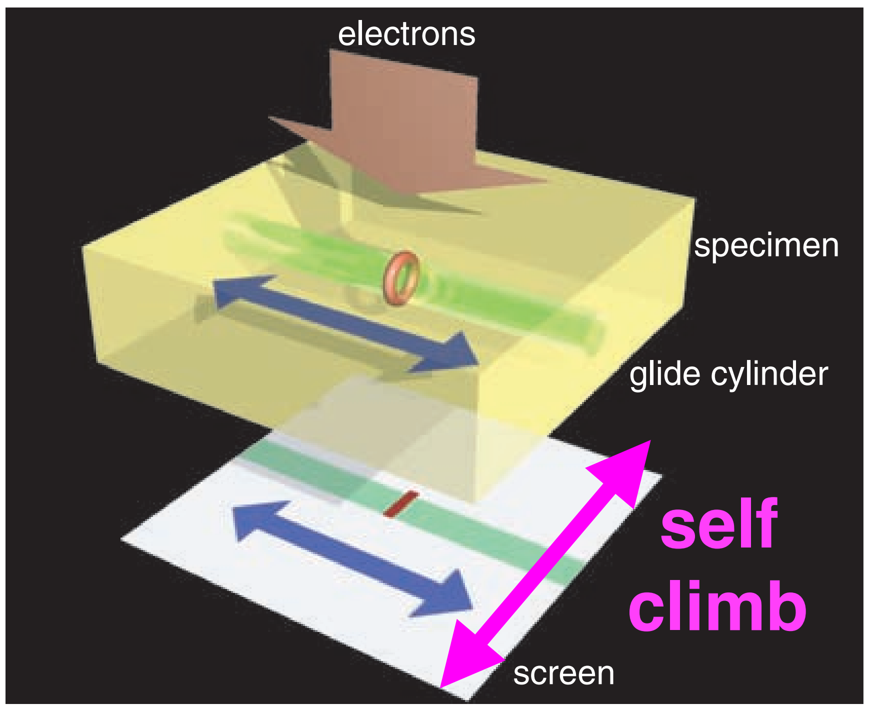

- $D_{SC}$ is **unchanged** when nucleation from flat surfaces is explicitly forbidden

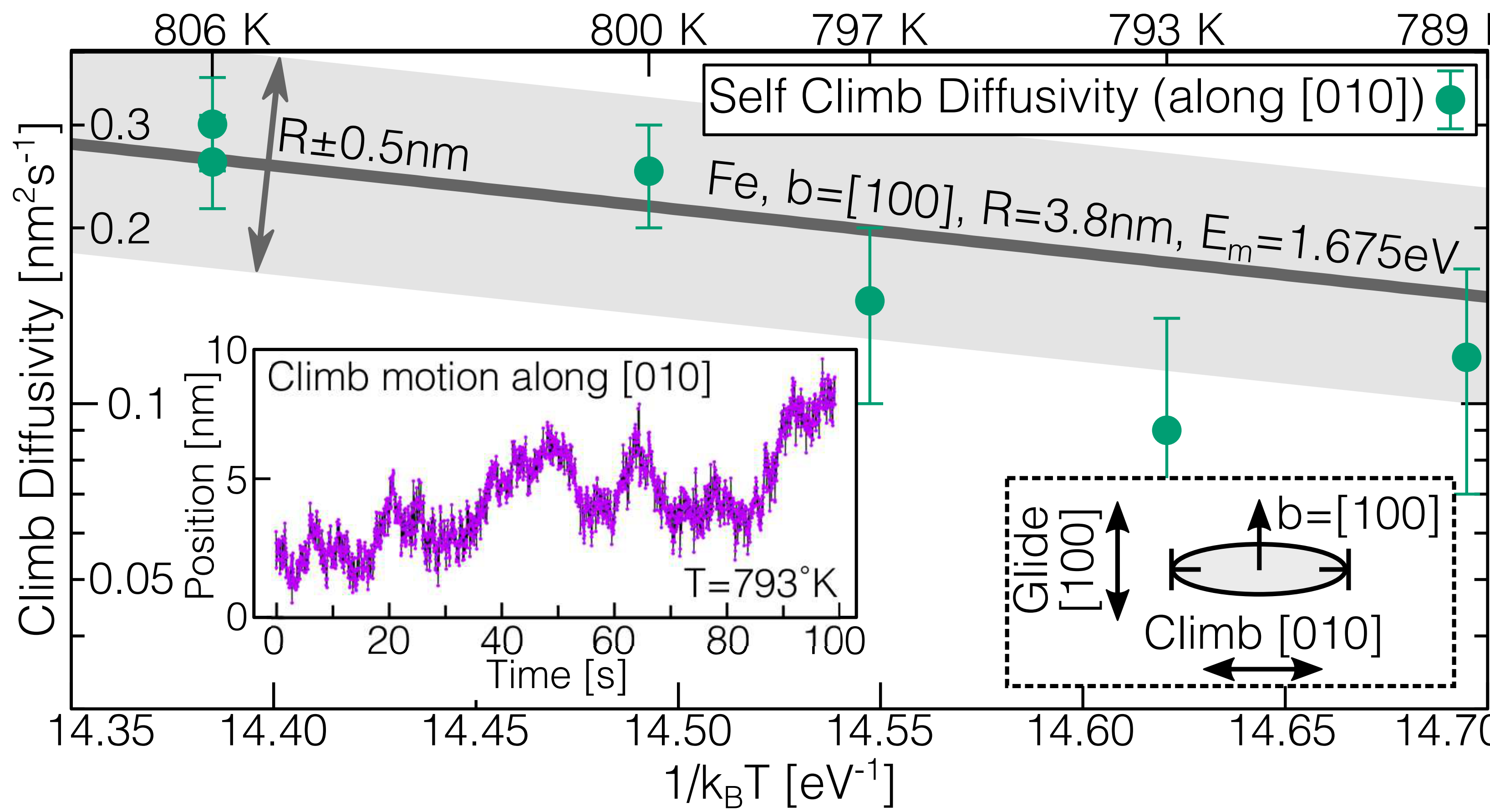
- Self climb is driven by the **intrinsic roughness** of `non-magic' number loops

TDS, K Arakawa, *et al.*, *Scientific Reports* 2016

# Unbiased self climb diffusion

- In our TEM experiments, essentially isolated prismatic loops were directly observed executing motion perpendicular to the loop Burgers vector



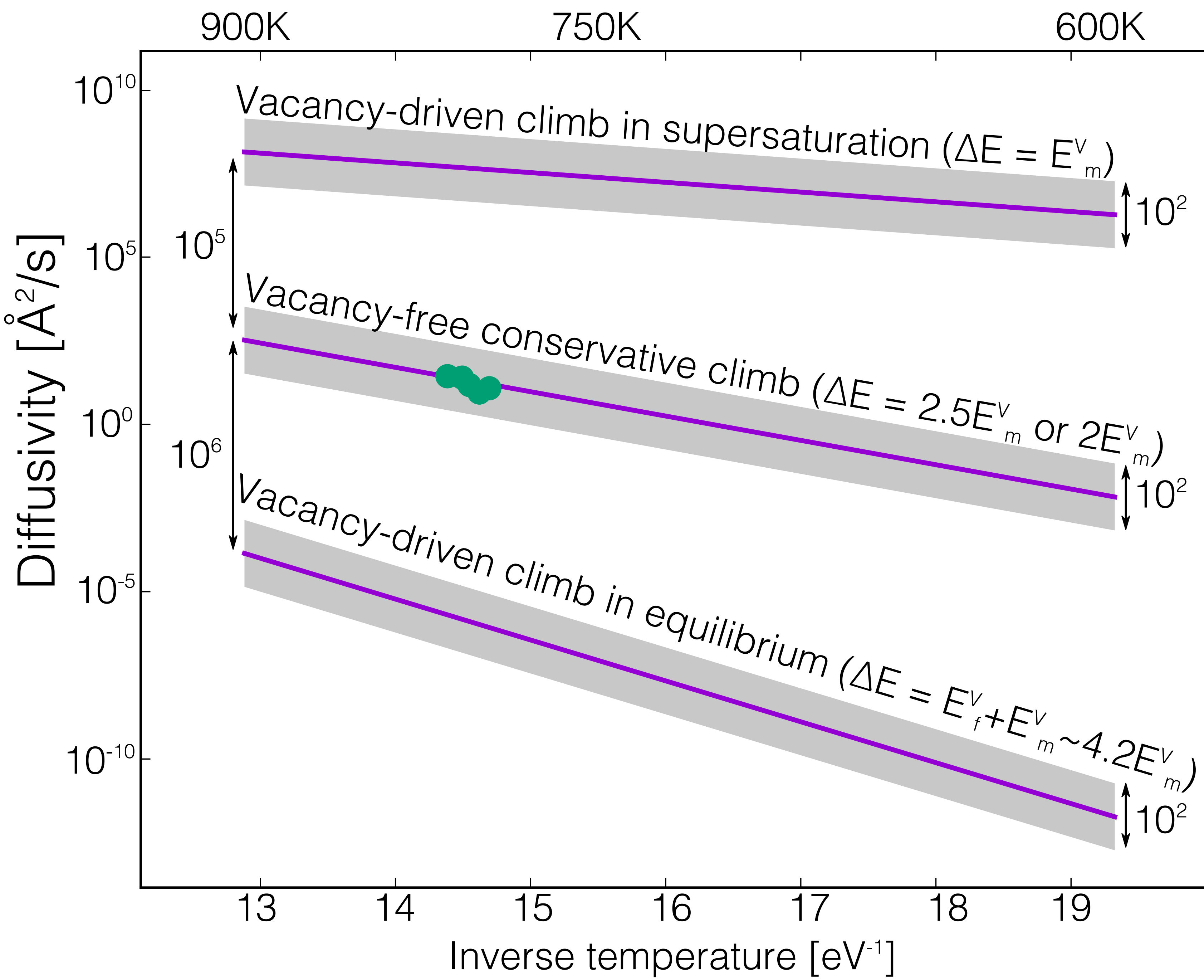

$$\mathrm{M_{scl}} = \frac{2\beta\nu a^5}{\pi R^3} e^{-\beta E_{\rm scl}} \qquad E^{\langle 100\rangle}_{\rm scl} = 2.5 E^V_m \qquad E_{\rm VMC} = 4.2 E^V_m$$

- Our calculated self climb mobility, (with $\nu_0$ from DFT, Sandberg *et al.* PRB 2015) gives good agreement with TEM measurements

TDS, K Arakawa, *et al., Scientific Reports* 2016

# Other possible mechanisms

- The only other candidates for such non-glide motion, even discarding the migration character, are either too fast or too slow




TDS, K Arakawa, *et al.*, *Scientific Reports* 2016

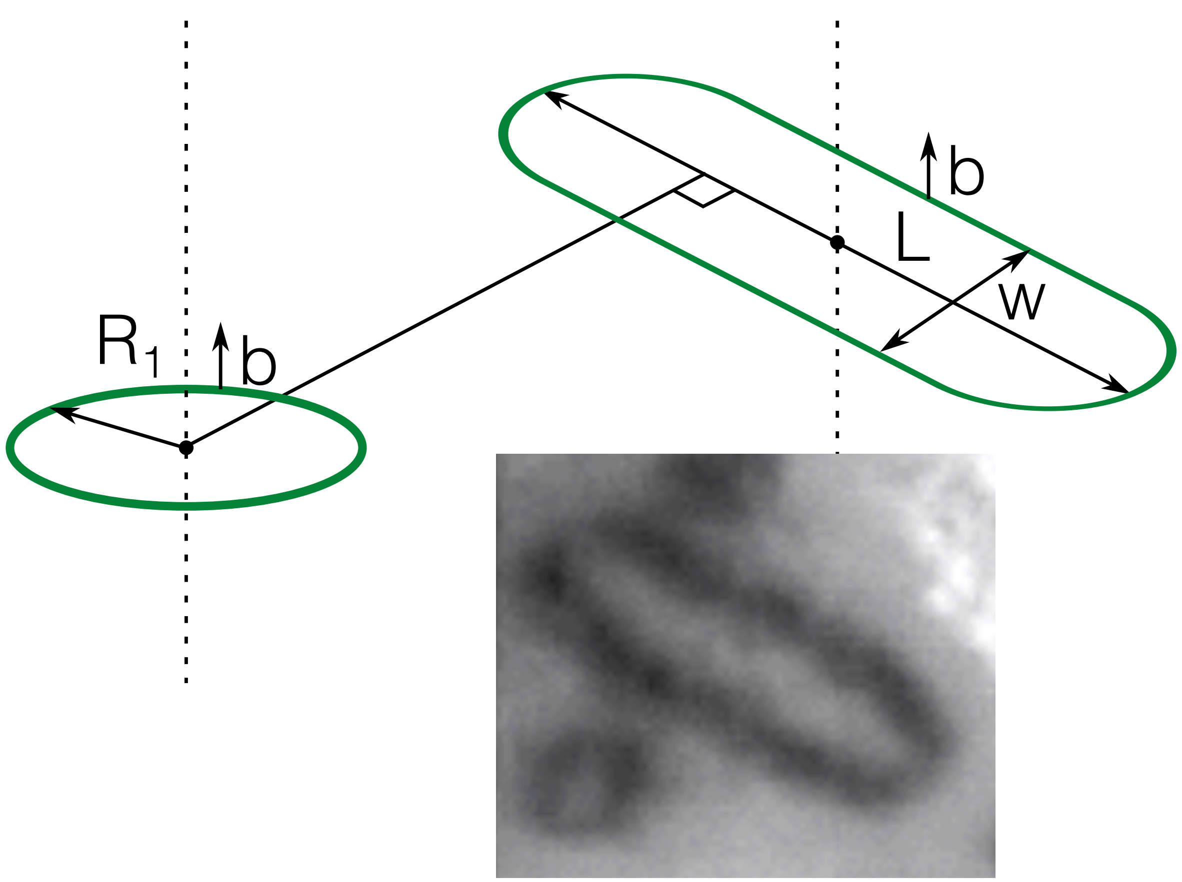
# Self climb in DD

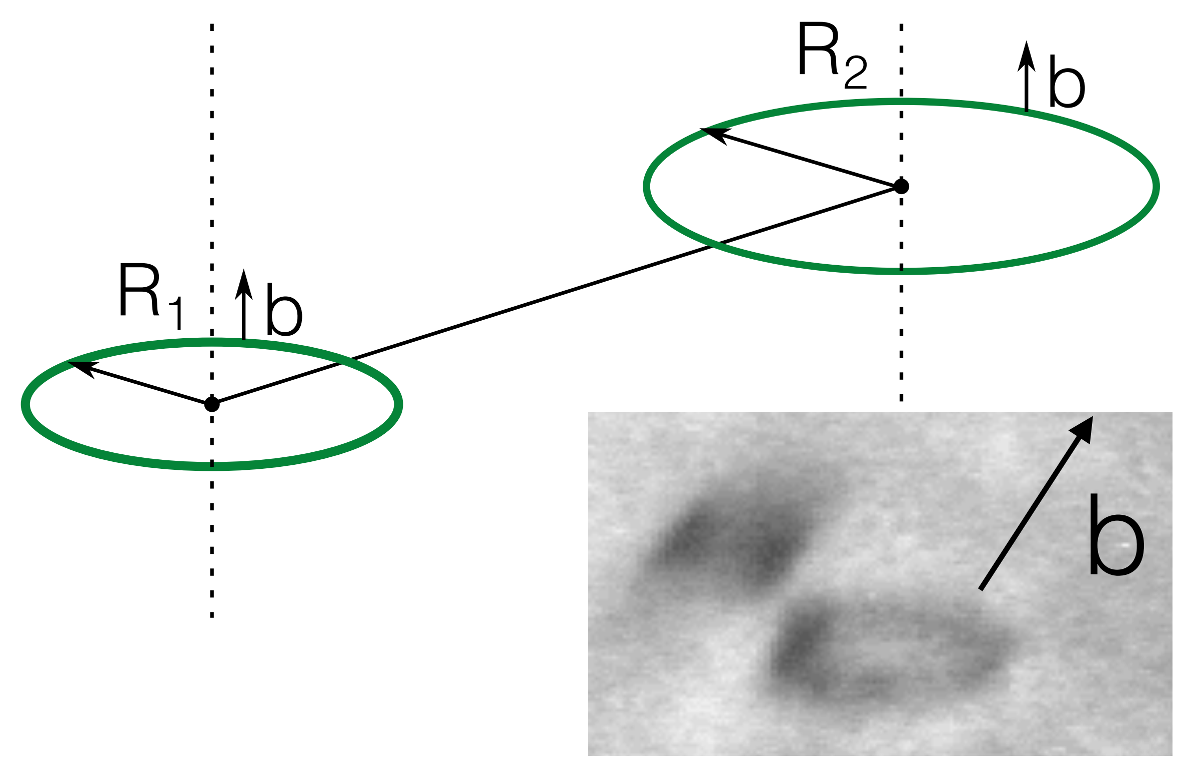
- In dislocation dynamics simulations allowing glide and self climb, we emulated binary loop coalescence processes seen under the TEM in Fe and W

$$W_{12} = \cancel{-\frac{\mu}{2\pi}\oint_{C_1}\oint_{C_2}\frac{(\mathbf{b}_1\times\mathbf{b}_2)\cdot(d\mathbf{l}_1\times d\mathbf{l}_2)}{R}} + \cancel{\frac{\mu}{4\pi}\oint_{C_1}\oint_{C_2}\frac{(\mathbf{b}_1\cdot d\mathbf{l}_1)(\mathbf{b}_2\cdot d\mathbf{l}_2)}{R}}$$

$$+\frac{\mu}{4\pi(1-\nu)}\oint_{C_1}\oint_{C_2}(\mathbf{b}_1\times d\mathbf{l}_1)\cdot\mathbf{T}\cdot(\mathbf{b}_2\times d\mathbf{l}_2) \qquad \text{(4-40)}$$

$$T_{ij} = \frac{\partial^2 R}{\partial x_i \partial x_j}$$




TDS, K Arakawa, *et al.*, *Scientific Reports* 2016

# Self climb in DD

- In dislocation dynamics simulations allowing glide and self climb, we emulated binary loop coalescence processes seen under the TEM in Fe and W

$$\mathbf{f}_{\mathrm{glide}} = -\hat{\mathbf{b}} \otimes \hat{\mathbf{b}} \cdot \nabla E_{\mathrm{elastic}} \qquad \mathrm{M}_{\mathrm{scl}} = \frac{2\beta\nu a^5}{\pi R^3} e^{-\beta E_{\mathrm{scl}}}$$

$$\mathbf{f}_{\mathrm{climb}} = -\left(1 - \hat{\mathbf{b}} \otimes \hat{\mathbf{b}}\right) \cdot \nabla E_{\mathrm{elastic}} \qquad \mathrm{M}_{\mathrm{glide}} \gg \mathrm{M}_{\mathrm{scl}}$$

$$\mathrm{v}_{\mathrm{glide}} = \mathrm{M}_{\mathrm{glide}} \mathrm{f}_{\mathrm{glide}} \qquad \mathbf{v}_{\mathrm{climb}} = \mathrm{M}_{\mathrm{scl}} \mathbf{f}_{\mathrm{climb}}$$

- As $\mathrm{v}_{\mathrm{glide}} \gg \mathrm{v}_{\mathrm{climb}}$ our simulation protocol is-
  - Let dislocations glide to a steady state due in some time $\delta t_{\mathrm{glide}}$
  - Calculate $\mathbf{f}_{\mathrm{climb}}$ and find $\delta t_{\mathrm{climb}}$ such that $\delta t_{\mathrm{climb}} |\mathbf{v}_{\mathrm{climb}}|^{\mathrm{max}} = (0.1 - 1)\text{Å}$
  - Update simulation time with $\delta t_{\mathrm{glide}} + \delta t_{\mathrm{climb}}$

TDS, K Arakawa, *et al.*, *Scientific Reports* 2016

# Comparison to experiment

- In dislocation dynamics simulations allowing glide and self climb, we emulated binary loop coalescence processes seen under the TEM in Fe and W

$$\mathbf{f}_{\text{glide}} = -\hat{\mathbf{b}} \otimes \hat{\mathbf{b}} \cdot \nabla E_{\text{elastic}}$$
$$\mathbf{f}_{\text{climb}} = -\left(1 - \hat{\mathbf{b}} \otimes \hat{\mathbf{b}}\right) \cdot \nabla E_{\text{elastic}}$$

$$\mathrm{M}_{\text{scl}} = \frac{2\beta\nu a^5}{\pi R^3} e^{-\beta E_{\text{scl}}}$$

| | $R_1$[nm] | $R_2$[nm] | $d$[nm] | T[K] | $\tau_{\text{exp}}$[s] | $\tau_{\text{SC}}$[s] | $\tau_{\text{VMC}}$[s] |
|---|---|---|---|---|---|---|---|
| Fe | 20 | 12 | 30 | 750 | 30.0 | 7.5 | $3.3 \times 10^7$ |
| Fe | 3.5 | 3.5 | 7 | 660 | ~0.8 | 1.8 | $2.7 \times 10^7$ |
| Fe$ | ~5. | ~5. | ~10. | 725 | ~6. | 2.1 | $2.7 \times 10^7$ |
| W† | 20 | 20 | 100 | 1173 | 66.5 | 96.2 | $2.6 \times 10^7$ |
| W† | 100 | 500* | 100 | 1273 | 7. | 8.6 | $1.5 \times 10^5$ |

- Hard to find examples that can be modelled with the simplistic dislocation geometry used in simulations but much closer than VMC

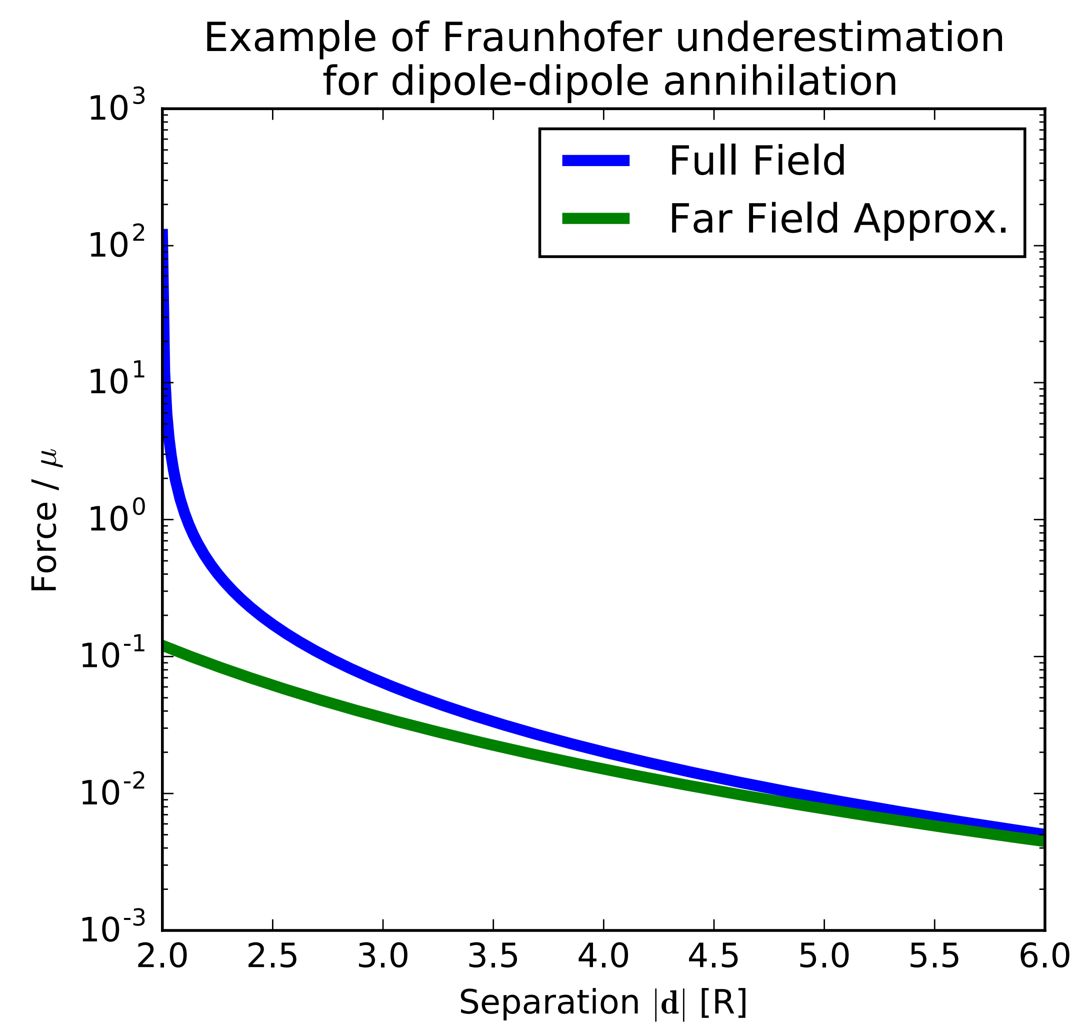
TDS, K Arakawa, *et al.*, *Scientific Reports* 2016

# Self climb in DD

- We also gauged the influence of self climb in simple DD simulations of post irradiation annealing of a box of ~80 prismatic 1/2<111> loops in Fe at 750K

- For computational simplicity we used the far field approximation (R << d) for parallel loop-loop interaction

$$-\nabla E^{12}_{\mathrm{elastic}} = \frac{\mu b^2 \pi^2 R_0^2 R_1^2}{4\pi(1-\nu)|\mathbf{d}|^4}\left(g(\theta)\hat{\mathbf{d}} - f(\theta)\hat{\mathbf{b}}\right)$$




- This approximation does not capture the force divergence and hence **underestimates** self-climb driven coalescence rates.

TDS, K Arakawa, *et al.*, *Scientific Reports* 2016

# Self climb in DD

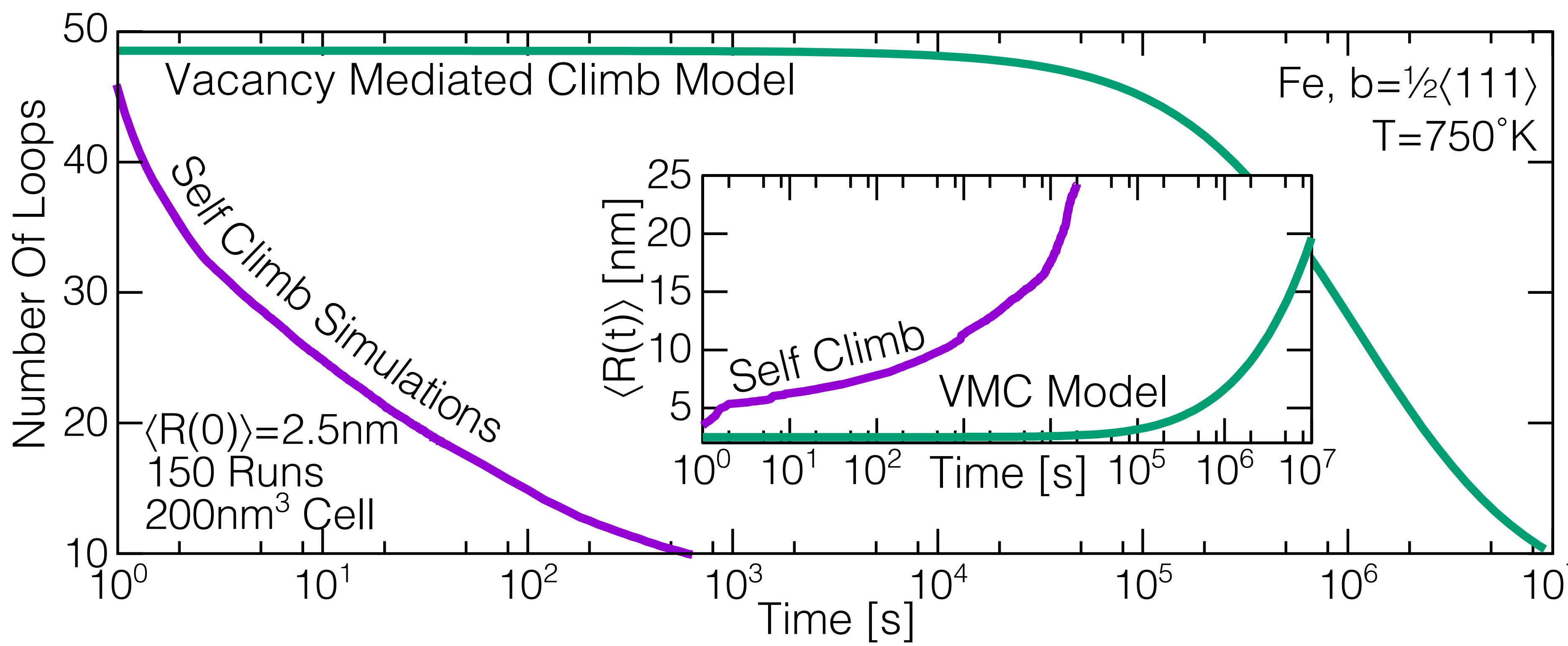
- We also gauged the influence of self climb in simple DD simulations of post irradiation annealing of a box of ~80 prismatic 1/2<111> loops in Fe at 750K

- Compared against an analytical model of vacancy mediated climb driven annealing, which has been successfully tested with VMC climb simulations

$$N_{\mathrm{loops}}(t) = \frac{N_{\mathrm{loops}}(0)}{1+\alpha t} \qquad \alpha = \frac{\beta\mu\Omega}{\langle R^2_{t=0}\rangle} e^{-\beta(E_{\mathrm{formation}}+E_{\mathrm{migration}})}$$

- Bakó, Clouet, Dupuy and Blétry Phil. Mag. 2011

- In agreement with experiment, self climb significantly affects PI annealing rates




TDS, K Arakawa, *et al.*, *Scientific Reports* 2016

# Outlook / Conclusions

- By considering structural fluctuations on timescales too great for MD, anomalously fast defect coalescence can be accurately modelled.
  TDS, K Arakawa, *et al.*, *Scientific Reports* 2016

- Self climb is fast due to the significantly lower activation energy

  Self-Climb: $E_{\mathrm{SC}} = (2\,\mathrm{or}\,2.5)E^V_m$ $E^V_f + E^V_m \simeq 4E^V_m$ (Fe)

  Climb: $E^V_m + E^V_f \simeq (3-5)E^V_m$ $E^V_f + E^V_m \simeq 3E^V_m$ (W)

- Consistent with previous experiments that found $E_{\mathrm{SC}} = (0.4 - 0.7)(E^V_f + E^V_m)$

- Inclusion of self climb mobilities, benchmarked against experiments, has a significant influence on the rate of post-irradiation annealing simulations

- Future work will investigate the proven role of self climb in post-irradiation annealing and explore applications to climb motion of dislocation lines

**Thank you for listening**

tomswinburne@gmail.com

tiny.cc/tds110

CCFE CULHAM CENTRE FOR FUSION ENERGY

EUROfusion