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

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Microstructure Evolution

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

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- The dominant change in microstructure is the growth of prismatic dislocation loops.

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 $\frac{1}{2}\langle 111 \rangle$ loops.

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

- Theory of **vacancy mediated climb (VMC)** gives
  \[ \text{VVMC} \propto C_{\text{vacancies}} \times V_{\text{ vacancy}} \]
  \[ \propto e^{-\beta (E_{\text{formation}} + E_{\text{migration}})} \]
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 $10^6$ faster than the predictions of climb theory.

$$V_{\text{VMC}} \propto e^{-\beta(E_{\text{formation}} + E_{\text{migration}})}$$

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

In situ high temperature isochronal (1 h, 800–1400°C) experiments with 1 nm separation would coalesce solely by vacancy-mediated climb in $\sim$1 h, making this hypothesis very unlikely based on the experimental observations reported here.

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
Climb in post irradiation annealing

<table>
<thead>
<tr>
<th>Feature</th>
<th>Vacancy Mediated Climb (VMC)</th>
<th>Experimental Observations</th>
</tr>
</thead>
<tbody>
<tr>
<td>Loop growth mechanism</td>
<td>Growth by the evaporation of small loops due to non-local vacancy flux balance</td>
<td>Direct coalescence by non-glide motion, with no change in loop area</td>
</tr>
<tr>
<td>Rate of climb motion</td>
<td>$\sim10^{-7}\text{Ås}^{-1}$ at $\sim0.3T_m$</td>
<td>$\sim10^{0.1}\text{Ås}^{-1}$ at $\sim0.3T_m$</td>
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</tbody>
</table>

- 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.
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 -

\[ \bar{x} = \sum_{i} \frac{x_i}{N}, \quad D_{SC} = \lim_{t \to \infty} \frac{\langle \bar{x}^2 \rangle}{2t} = \frac{N_P}{N^2} D_P \]

\[ \frac{v_{climb}}{f_{climb}} \equiv M_{SC} = \beta D_{SC} = \frac{2\beta a^5}{\pi R^3} \nu_0 e^{-\beta E_{SC}} \]

• 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

\[ E_{SC} \sim (0.4 - 0.7)(E_{\text{formation}} + E_{\text{migration}}) \]
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!
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)
Climb without vacancies

- All observed pathways could clearly be split into jumps along primitive lattice vectors $1/2\langle 111 \rangle$, $\langle 111 \rangle$, $1/2\langle 111 \rangle$

- The energy barriers in Fe,W could be normalised by the vacancy migration barrier
Climb without vacancies

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

\[ \frac{1}{2}\langle 111 \rangle : \Delta E \simeq \Delta c E_m^V, \quad E_m \simeq (\Delta c + 1) E_m^V \]
\[ <100> : \Delta E \simeq \Delta c E_m^V, \quad E_m \simeq (\Delta c + 2) E_m^V \]

\( E_m^V \): vacancy migration barrier
\( \Delta c \): change in number of SIA neighbours

\[ c_i = \sum_{j \neq i} \Theta \left( \frac{\sqrt{3}}{2} a - |x_i - x_j| \right) \]

- 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
Climb without vacancies

\[ \Delta E = \Delta c E_m^V \]
\[ E_m^{(111)/2} = (\Delta c + 1) E_m^V \]
\[ E_m^{(100)/2} = (\Delta c + 2) E_m^V \]

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

\[ E_scl^{(100)} = 2.5 E_m^V, \quad E_scl^{1/2(111)} = 2.0 E_m^V \]

\[ M_{scl} = \frac{2\beta \nu a^5}{\pi R^3} e^{-\beta E_{scl}} \]
Climb without vacancies

\[ E_{\text{scl}}^{(100)} = 2.5E_m^V, \quad E_{\text{scl}}^{1/2(111)} = 2.0E_m^V \]

- The activation energy corresponds to SIA migration around loop corners

\[ \Delta c = 0 \quad (1) \quad \langle E_m \rangle \approx 2E_m^V \quad \langle \Delta E \rangle \approx 0 \]

\[ \Delta c = 3 \quad \langle E_m \rangle \approx 4E_m^V \quad \langle \Delta E \rangle \approx 3E_m^V \]

- \( D_{\text{SC}} \) 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.

\[
M_{\text{scl}} = \frac{2\beta v a^5}{\pi R^3} e^{-\beta E_{\text{scl}}} \quad \quad \quad E_{\text{scl}}^{(100)} = 2.5 E_m^V \quad \quad \quad E_{\text{VMC}} = 4.2 E_m^V
\]

- Our calculated self climb mobility, (with \( v_0 \) from DFT, Sandberg et al. PRB 2015) gives good agreement with TEM measurements.
Other possible mechanisms

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

\[
W_{12} = -\frac{\mu}{2\pi} \int_{C_1} \int_{C_2} \frac{b_1 \times b_2 \cdot (d_1 \times d_2)}{R} + \frac{\mu}{4\pi} \int_{C_1} \int_{C_2} \frac{(b_1 \cdot d_1)(b_2 \cdot d_2)}{R}
\]

\[
+ \frac{\mu}{4\pi(1-\nu)} \int_{C_1} \int_{C_2} (b_1 \times d_1) \cdot T \cdot (b_2 \times d_2)
\]

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

(4-40)
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

\[
\mathbf{f}_{\text{glide}} = -\mathbf{b} \otimes \mathbf{b} \cdot \nabla E_{\text{elastic}} \quad \mathbf{M}_{\text{scl}} = \frac{2\beta \nu a^5}{\pi R^3} e^{-\beta E_{\text{scl}}}
\]

\[
\mathbf{f}_{\text{climb}} = -\left(1 - \mathbf{b} \otimes \mathbf{b}\right) \cdot \nabla E_{\text{elastic}} \quad \mathbf{M}_{\text{glide}} \gg \mathbf{M}_{\text{scl}}
\]

\[
\mathbf{v}_{\text{glide}} = \mathbf{M}_{\text{glide}} \mathbf{f}_{\text{glide}} \quad \mathbf{v}_{\text{climb}} = \mathbf{M}_{\text{scl}} \mathbf{f}_{\text{climb}}
\]

- As \(\mathbf{v}_{\text{glide}} \gg \mathbf{v}_{\text{climb}}\) our simulation protocol is-

  - Let dislocations glide to a steady state due in some time \(\delta t_{\text{glide}}\)

  - Calculate \(\mathbf{f}_{\text{climb}}\) and find \(\delta t_{\text{climb}}\) such that \(\delta t_{\text{climb}} |\mathbf{v}_{\text{climb}}|_{\text{max}} = (0.1 - 1)\text{Å}\)

  - Update simulation time with \(\delta t_{\text{glide}} + \delta t_{\text{climb}}\)
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

\[ f_{\text{glide}} = -\hat{b} \otimes \hat{b} \cdot \nabla E_{\text{elastic}} \]

\[ f_{\text{climb}} = -\left(1 - \hat{b} \otimes \hat{b}\right) \cdot \nabla E_{\text{elastic}} \]

\[ M_{\text{scl}} = \frac{2\beta\nu a^5}{\pi R^3} e^{-\beta E_{\text{scl}}} \]

<table>
<thead>
<tr>
<th></th>
<th>(R_1[\text{nm}])</th>
<th>(R_2[\text{nm}])</th>
<th>(d[\text{nm}])</th>
<th>(T[\text{K}])</th>
<th>(\tau_{\text{exp}}[\text{s}])</th>
<th>(\tau_{\text{sc}}[\text{s}])</th>
<th>(\tau_{\text{VMC}}[\text{s}])</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe</td>
<td>20</td>
<td>12</td>
<td>30</td>
<td>750</td>
<td>30.0</td>
<td>7.5</td>
<td>(3.3 \times 10^7)</td>
</tr>
<tr>
<td>Fe</td>
<td>3.5</td>
<td>3.5</td>
<td>7</td>
<td>660</td>
<td>(~0.8)</td>
<td>1.8</td>
<td>(2.7 \times 10^7)</td>
</tr>
<tr>
<td>Fe (^\dagger)</td>
<td>(~5.)</td>
<td>(~5.)</td>
<td>(~10.)</td>
<td>725</td>
<td>(~6.)</td>
<td>2.1</td>
<td>(2.7 \times 10^7)</td>
</tr>
<tr>
<td>W (^\dagger)</td>
<td>20</td>
<td>20</td>
<td>100</td>
<td>1173</td>
<td>66.5</td>
<td>96.2</td>
<td>(2.6 \times 10^7)</td>
</tr>
<tr>
<td>W (^\dagger)</td>
<td>100</td>
<td>500*</td>
<td>100</td>
<td>1273</td>
<td>7.</td>
<td>8.6</td>
<td>(1.5 \times 10^5)</td>
</tr>
</tbody>
</table>

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

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

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

\[ N_{\text{loops}}(t) = \frac{N_{\text{loops}}(0)}{1 + \alpha t} \quad \alpha = \frac{\beta \mu \Omega}{\langle R_t^2 \rangle} e^{-\beta(E_{\text{formation}} + E_{\text{migration}})} \]

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

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

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**Figure 2**

Vacancy Mediated Climb Model

- Fe, b=½(111)
- T=750°C

- \( \langle R(0) \rangle = 2.5 \text{nm} \)
- 150 Runs
- 200nm³ Cell

- Self Climb
- VMC Model

**Figure 3**

Evolution of the loop average radius observed in our DD simulations with the one predicted by Equation (13). Figure 2 shows that quantitative agreement is obtained.
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: $E_{SC} = (2 \text{ or } 2.5)E_m^V$ \hspace{1cm} $E_f^V + E_m^V \approx 4E_m^V$ (Fe)  
  
  Climb: $E_m^V + E_f^V \approx (3 - 5)E_m^V$ \hspace{1cm} $E_f^V + E_m^V \approx 3E_m^V$ (W)

- Consistent with previous experiments that found $E_{SC} = (0.4 - 0.7)(E_f^V + E_m^V)$

- 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
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