In-situ TEM Study of the Effects of W Solutes on Irradiation Induced Detwinning in Cu

SAND2018-2351C

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Funded by US DOE – BES under grant DEFG02-05ER46217

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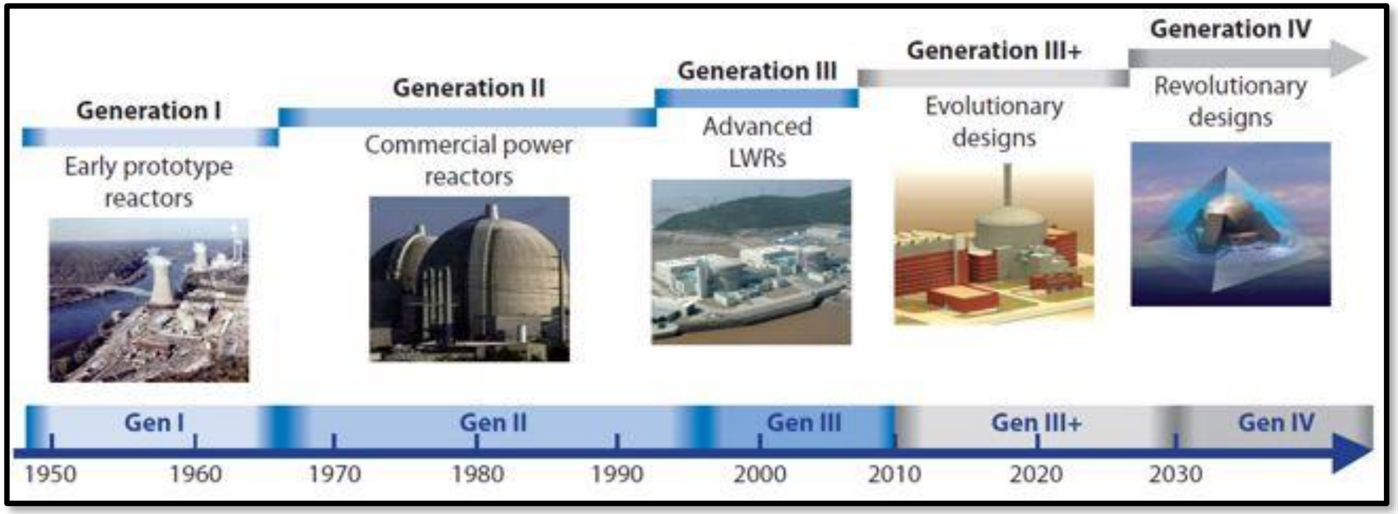
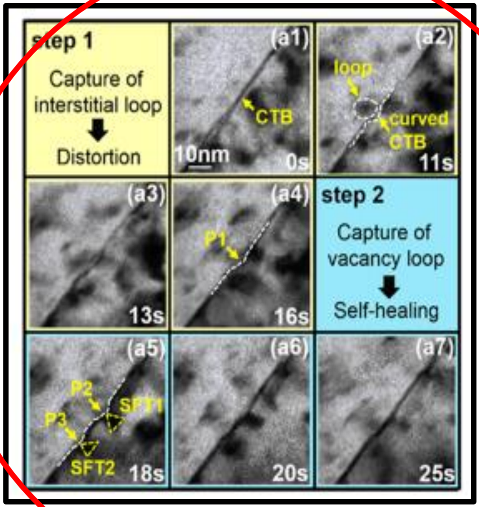
Nanostructure materials with high sink density as candidates for extreme irradiation and temperature environments



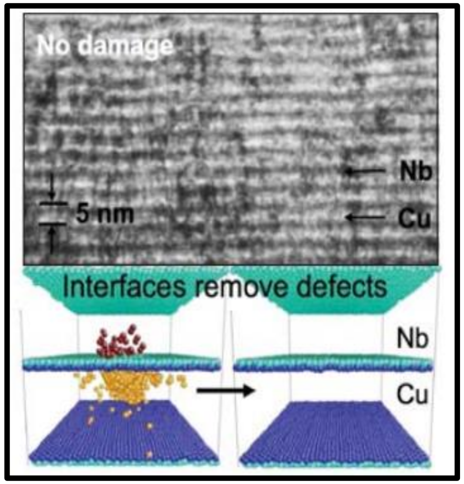
Need for materials that can last 50-100 years in extreme environments.

Nanotwinned (nt) metals



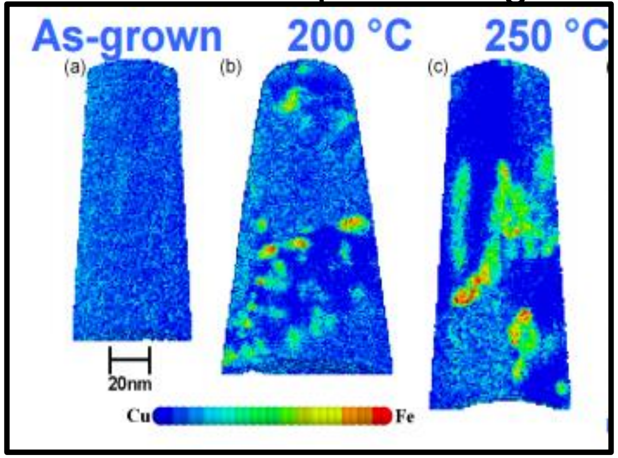
J. Li et. al. Nano Let.. 15 (2015).

Nanolaminates



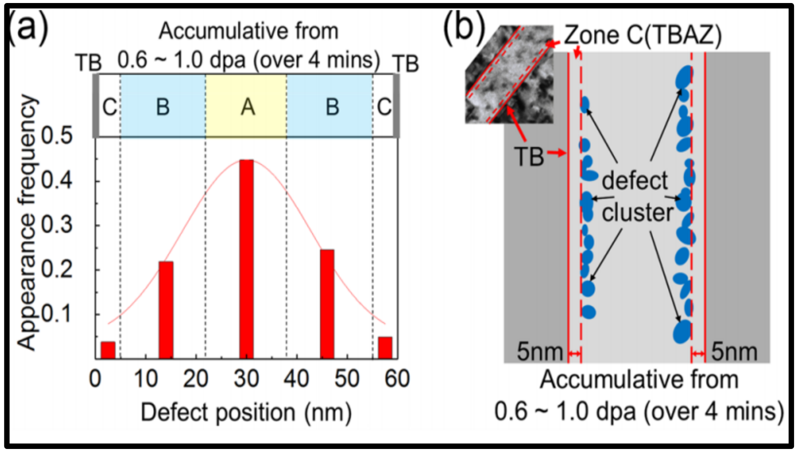
Zhang et. al. Nucl. Instrum. Methods Phys. Res., Sect. B., 261 (2007)

Nano-scale patterning



Chee et al. Acta Mater., 58 (2010)

High density of twins in nt metals reduce defect density but are prone to detwinning through interaction with defects



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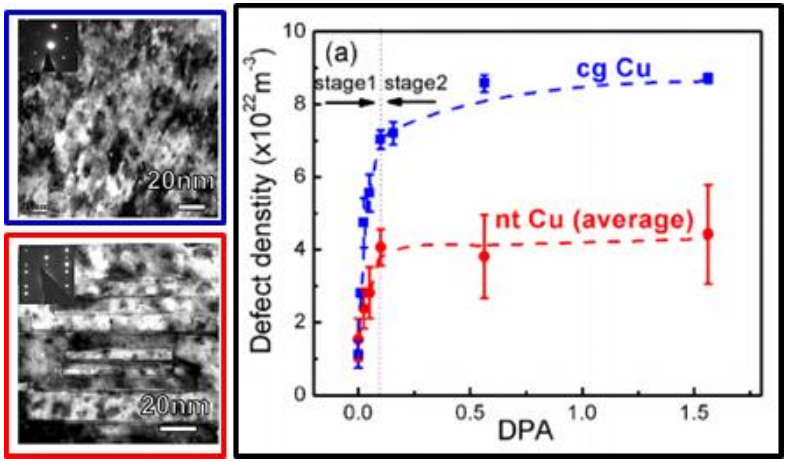
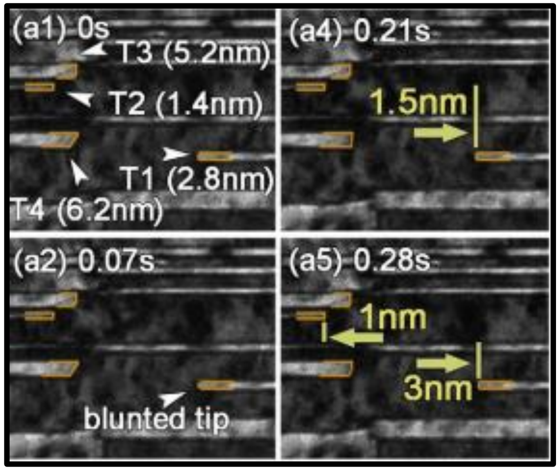
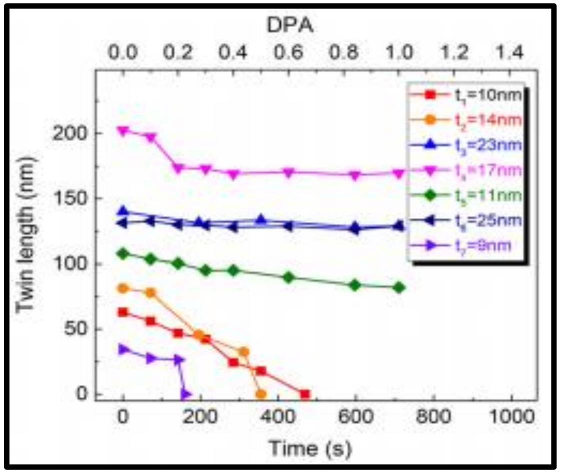


Y. Chen et. al. Acta Mater. 111 (2016).

Defect clusters have smaller lifetime in nt-metals compared to coarse samples.



Y. Chen et. al. Scr. Mater. 130 (2017).



C. Fan et. al. Metall. Mater. Trans. A 48 (2017).

Coherent Twin Boundaries→ High energy {111} and

Incoherent twin boundaries→ Migrate to reduce CTB energy.

Migration of ITBs →glide of component Shockley partials with $b = \frac{1}{6} < 1\bar{2}1 >$ through interaction with defects.

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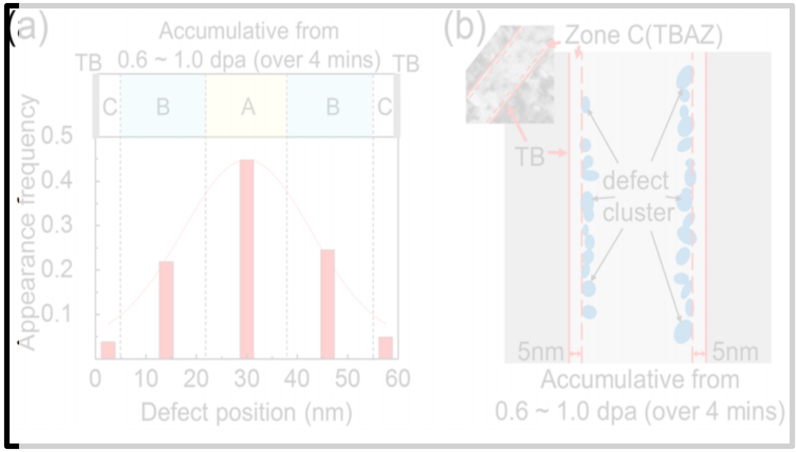
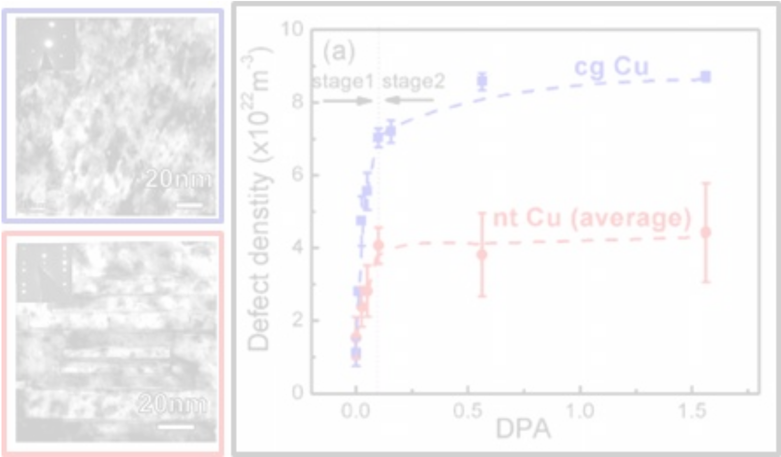


J. Li et. al. Nano Let.. 15 (2015).



Y. Chen et. al. Acta Mater. 111 (2016).

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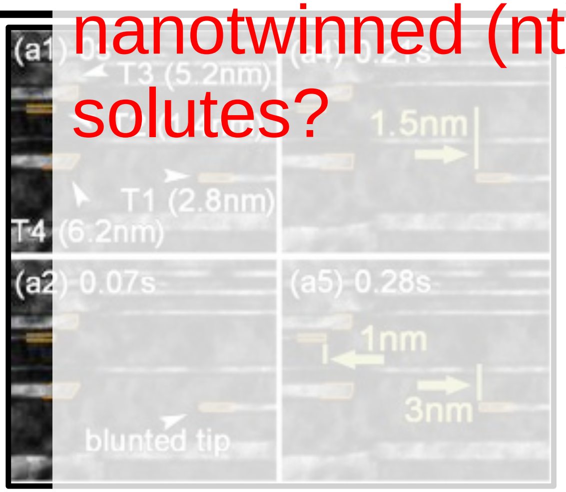
Is it possible to control or reduce detwinning in nanotwinned (nt) metals by alloying with immiscible solutes?



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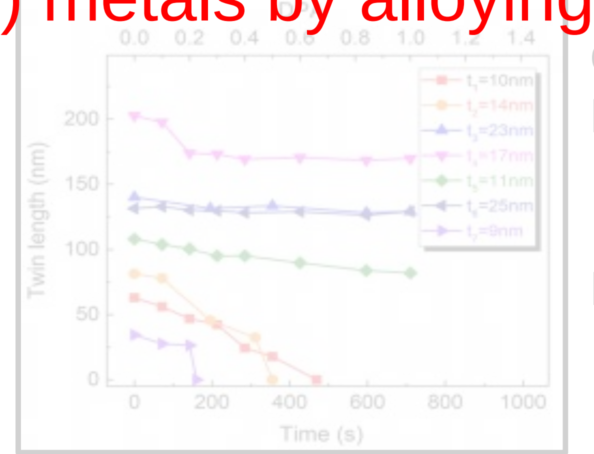

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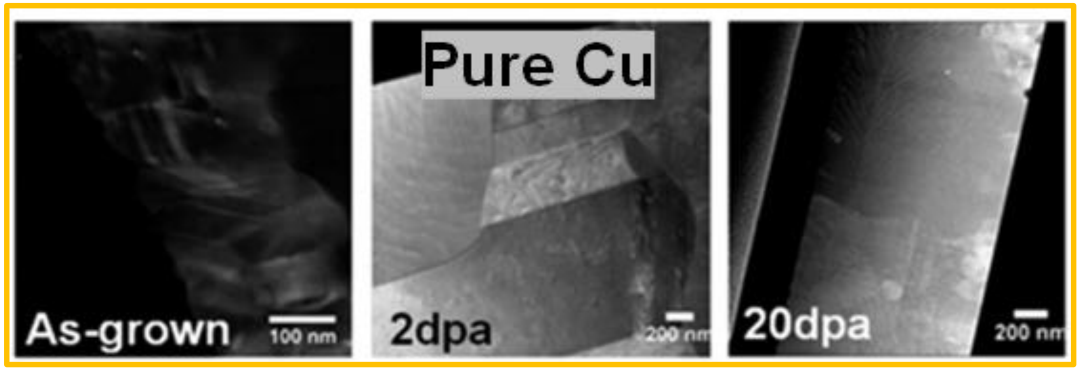
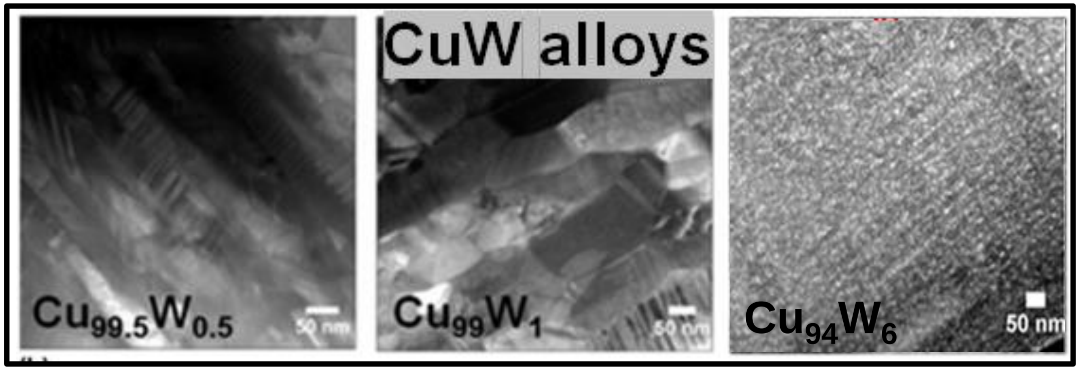
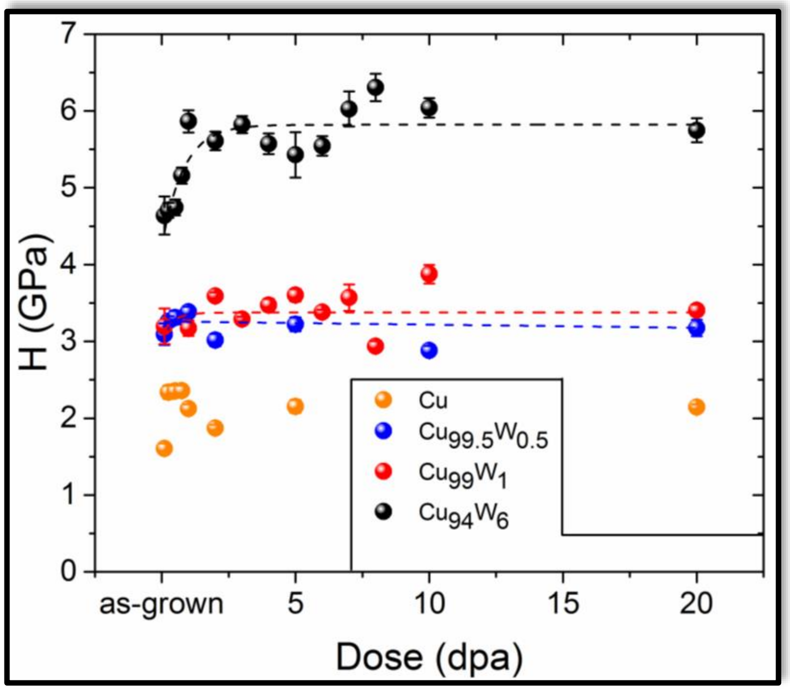
W solutes and precipitates in Cu stabilize nanotwins and stabilize their mechanical properties





G. Jawaharram et. al. J. Mater. Res. 32 (2017)

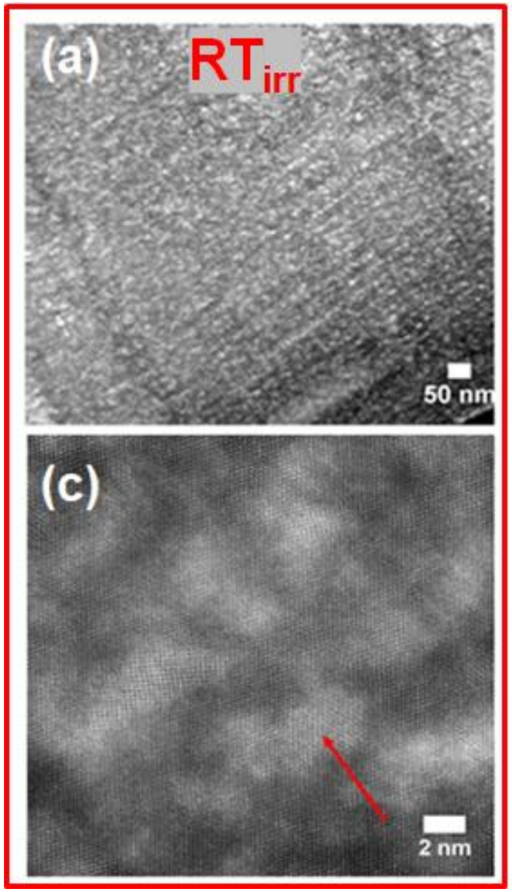


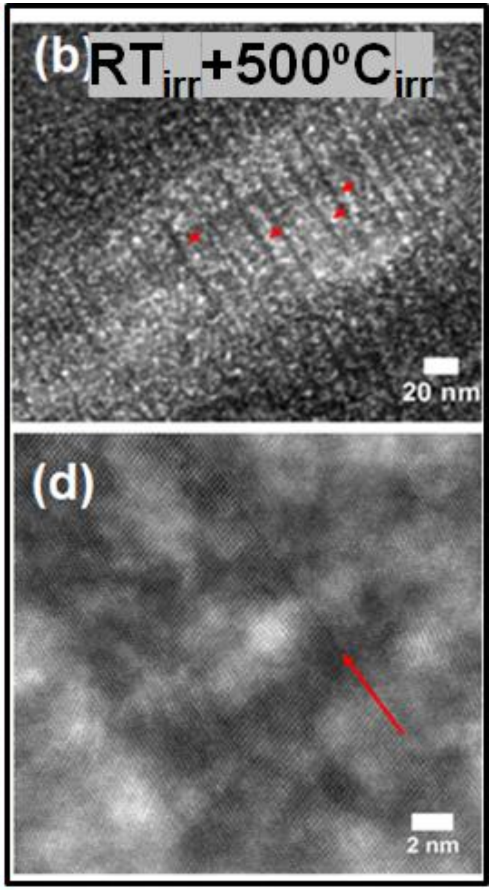
W solutes ($Cu_{99.5}W_{0.5}$) and precipitates ($Cu_{99}W_1$ and $Cu_{94}W_6$) stabilize nanotwins in Cu and prevent detwinning at doses as high as 20dpa.

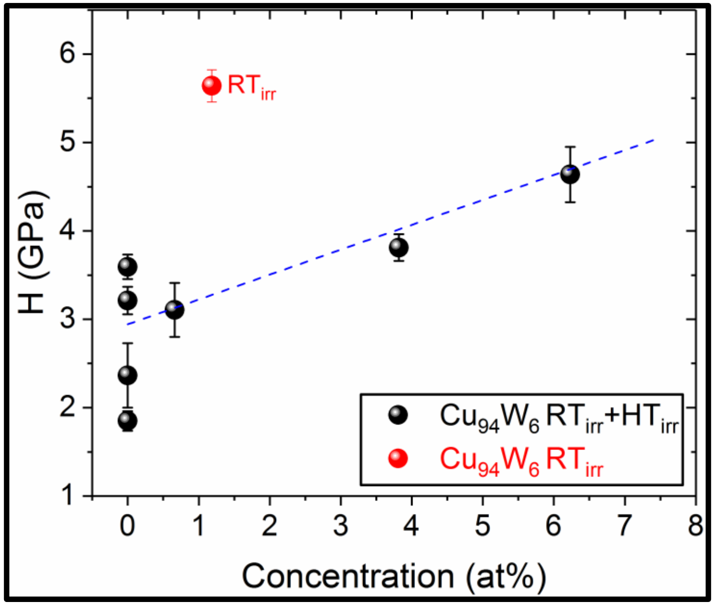
Average twin spacing decreases with increasing W concentration.

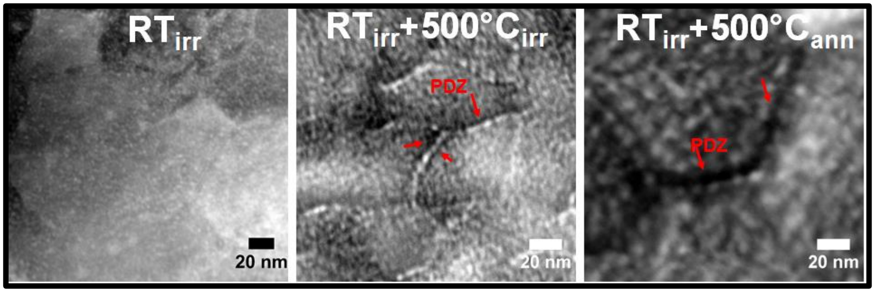
~250nm, 10.8nm, 9.7nm and 2.9nm for Cu, $Cu_{99.5}W_{0.5}$, $Cu_{99}W_1$ and $Cu_{94}W_6$.

Temperature affects the precipitate distribution around twins and grain boundaries and hence the mechanical properties









RT_{irr} → vacancy motion in Cu is negligible, and W particles can be trapped in thermodynamically unfavorable configurations due to the highly non-equilibrium precipitation mechanism.

HT_{irr}→ local thermodynamic relaxations are possible leading to a spatially non-uniform distribution of W particles around coherent twins in Cu.

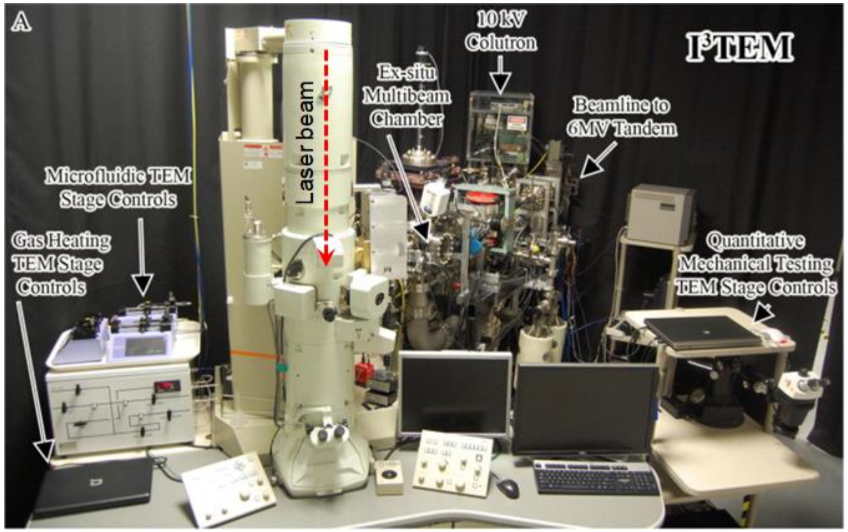
Sample preparation and in-situ TEM

All in-situ irradiation experiments were carried out using the I^{3}TEM at Sandia National Laboratories

1. Cu, $Cu_{99}W_1$ → synthesized using DC magnetron co-sputtering from Cu and W targets.

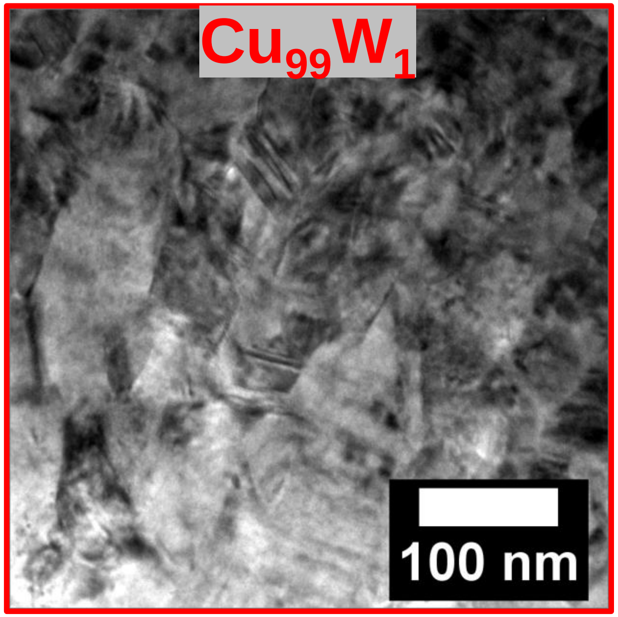
2. Irradiation parameters
   - Beam Energy – 2.6MeV Ag^{3+}
   - Ion Beam current – 63-68nA

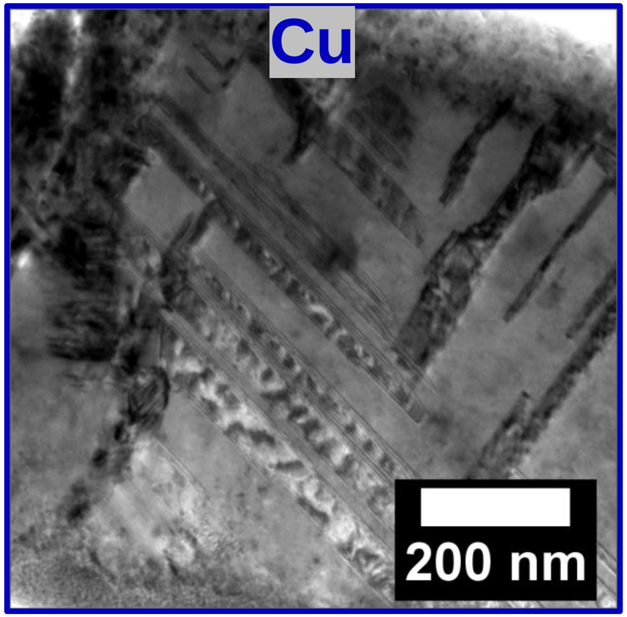
3. Heating source → 1064nm laser with a spot size of 50µm.

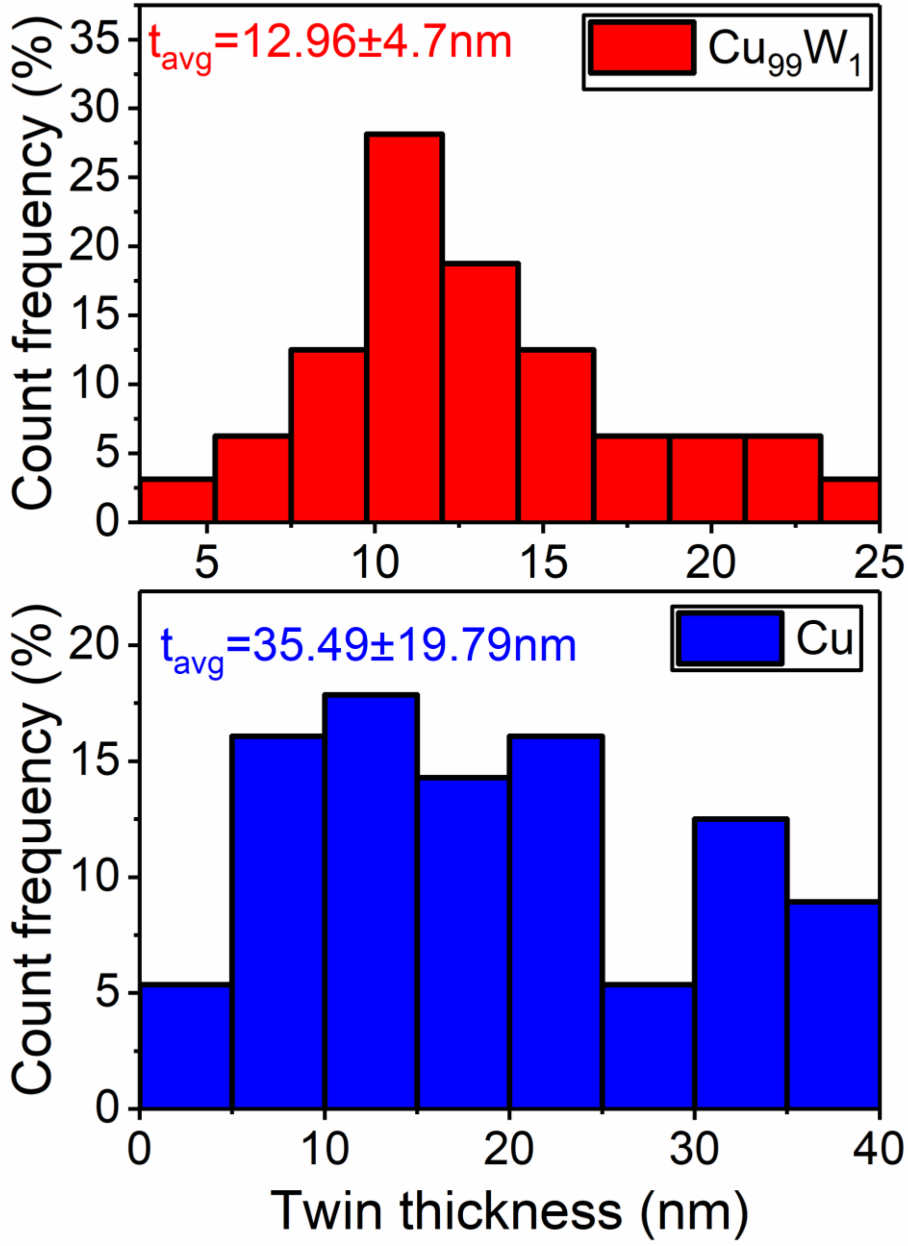


K. Hattar et. al. Nucl. Instr. Meth. Phys. Res. B 338 (2014).

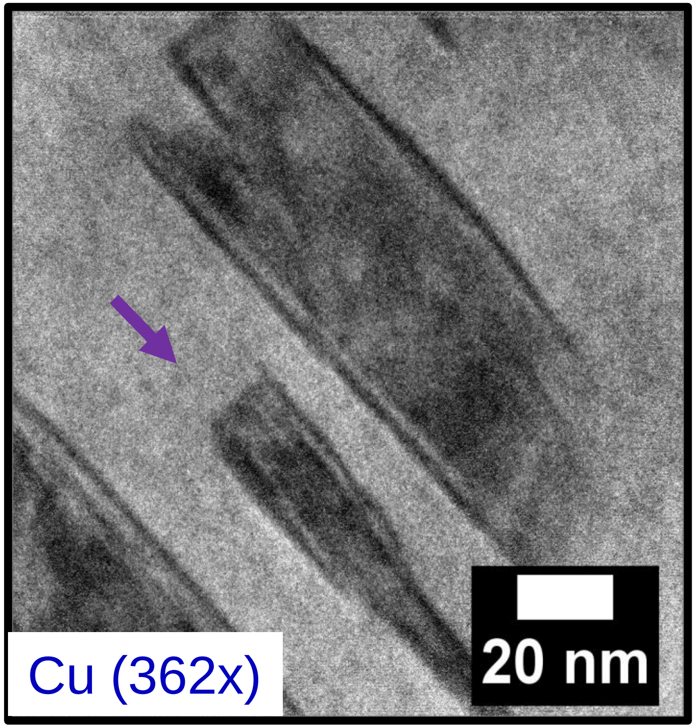
The initial twin thickness of $Cu_{99}W_1$ is smaller compared to pure Cu

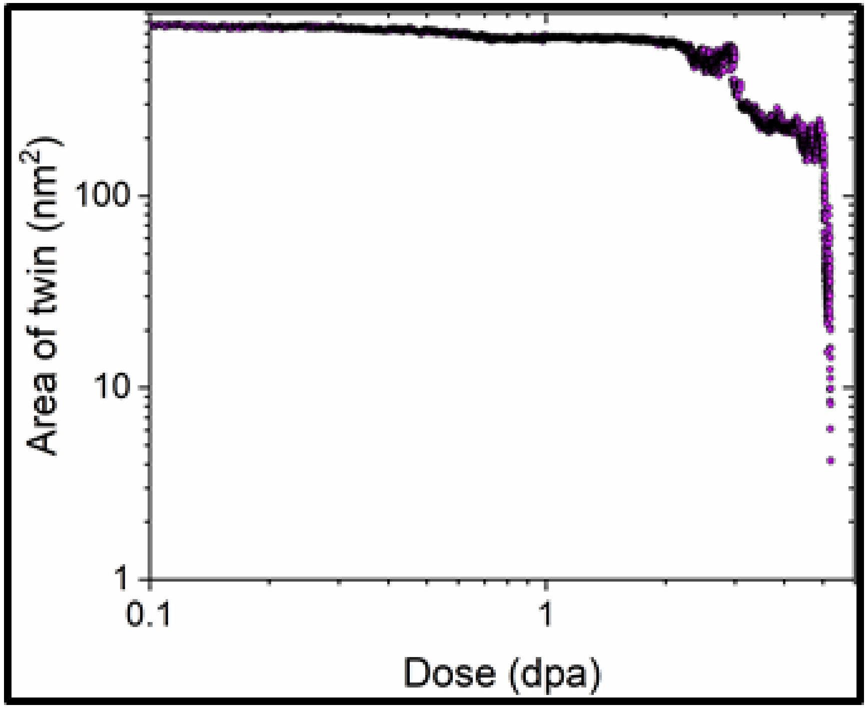






Nanotwins in pure Cu act as sinks for defect loops and undergo rapid detwinning through the migration of ITBs.





→ Detwinning is not continuous simultaneous growth and reduction of twin length.

→ Detwinning occurs through the migration of ITBs through absorption of defects and retwinning occurs through emission.

→ Defect loops interact CTB and ITBs which act as efficient sink sites.

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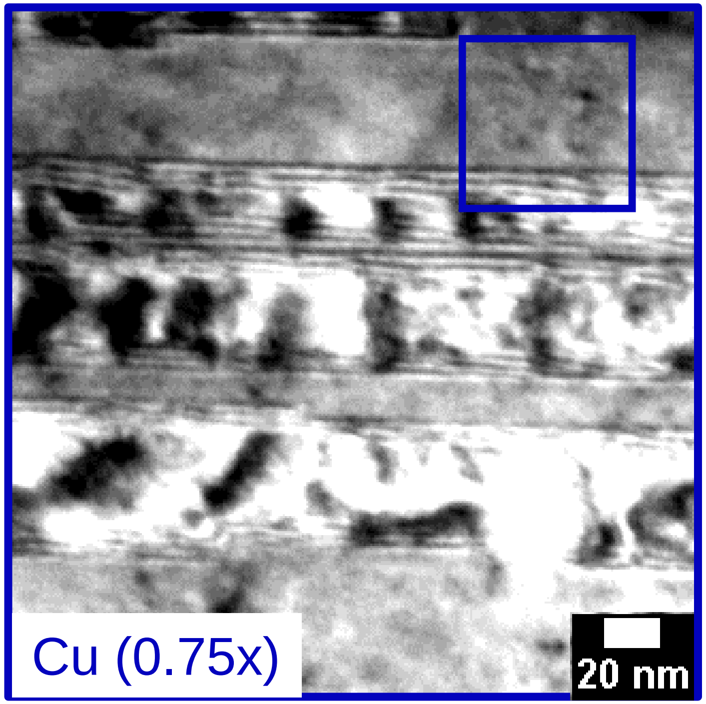

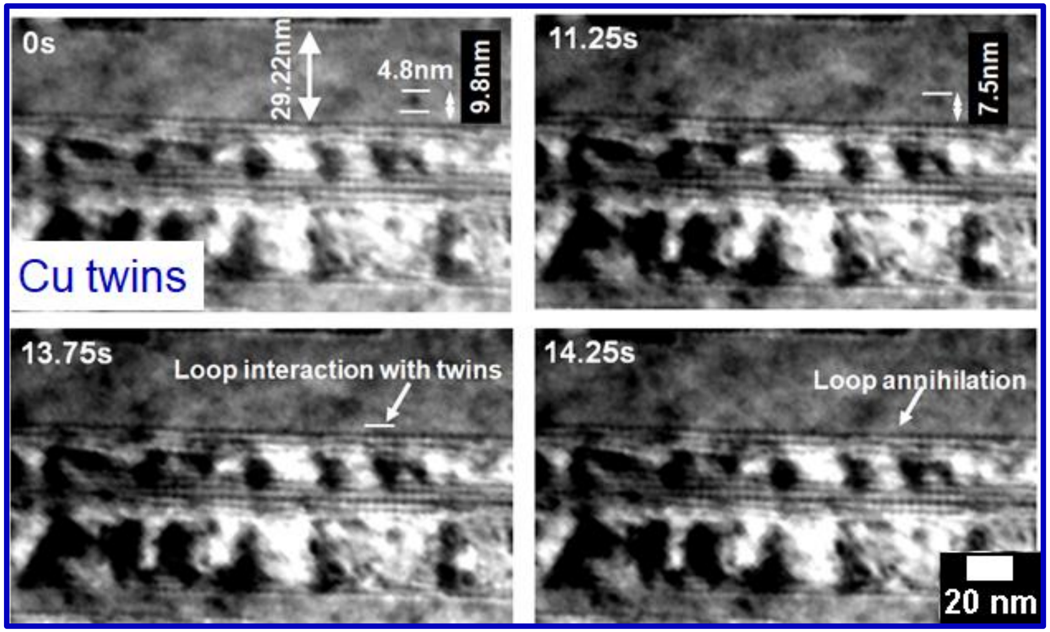

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C. Fan et. al. Metall. Mater. Trans. A 48 (2017).

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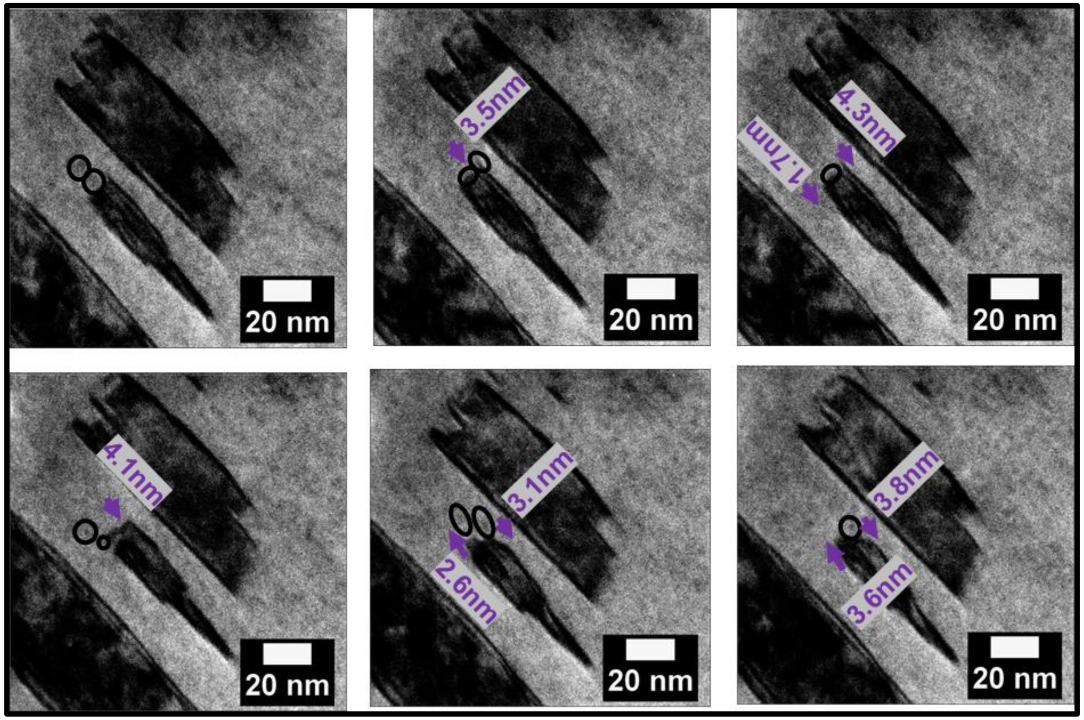
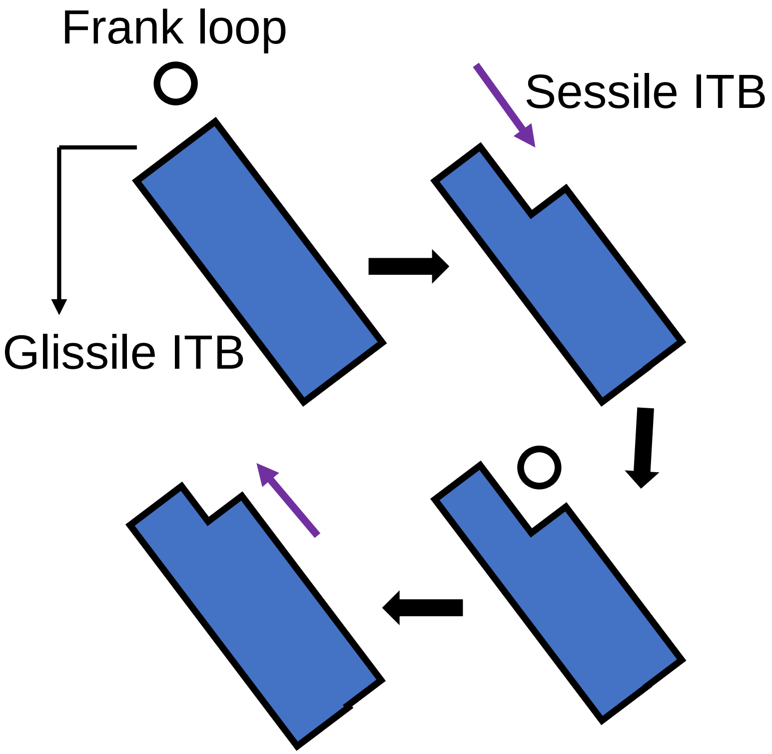




Detwinning

$$\frac{1}{2}[\bar{1}\bar{1}0]+\frac{1}{6}[112] \rightarrow \frac{1}{3}[\bar{1}\bar{1}1]$$

Frank loops

Shockley Partial (glissile)

Frank Partial (sessile)

Retwinning

$$\frac{1}{2}[110]+\frac{1}{3}[\bar{1}\bar{1}1] \rightarrow \frac{1}{6}[112]$$

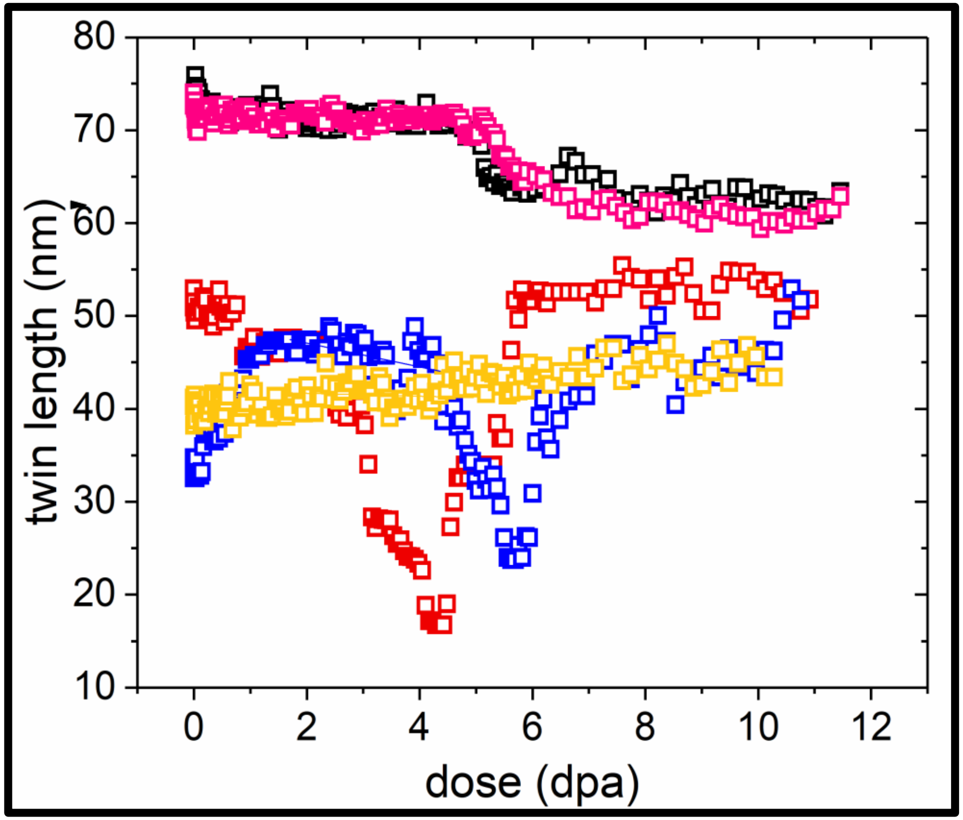
Frank Partial (sessile)

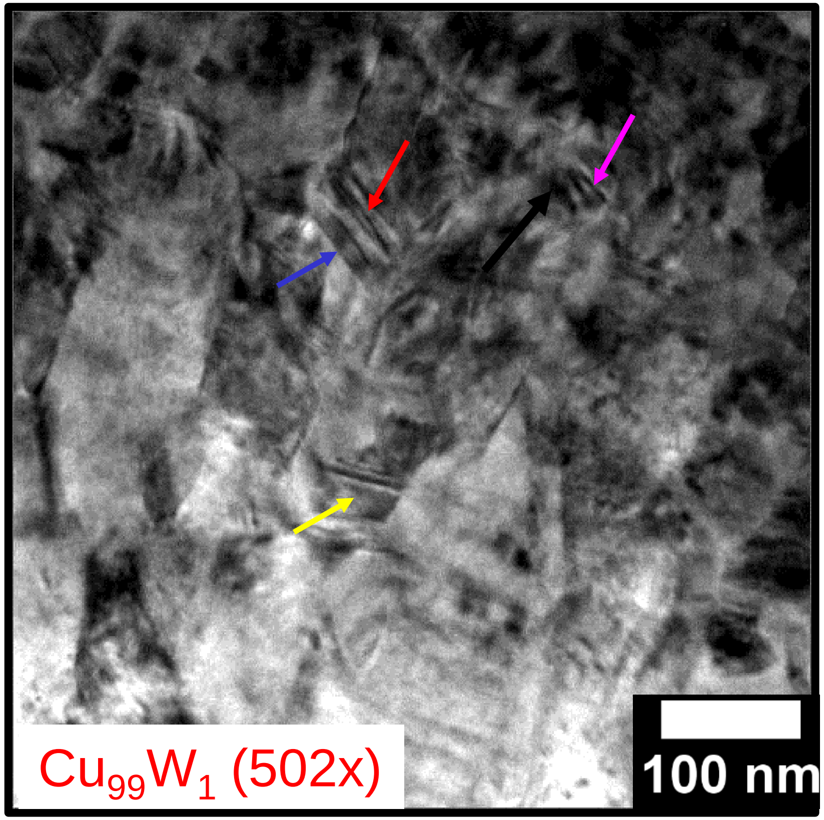
Frank loops

Shockley Partial (glissile)

C. Fan et. al. Metall. Mater. Trans. A 48 (2017).

Detwinning is extremely sluggish and occurs as a result of grain growth/shrinkage rather than ITB migration





→ITB nucleation is limited and the detwinning is insignificant compared to Cu.

→V – Shaped curve results from grain size rather than change in twin length through ITB migration.

→Twins tends to increase in length in certain cases.

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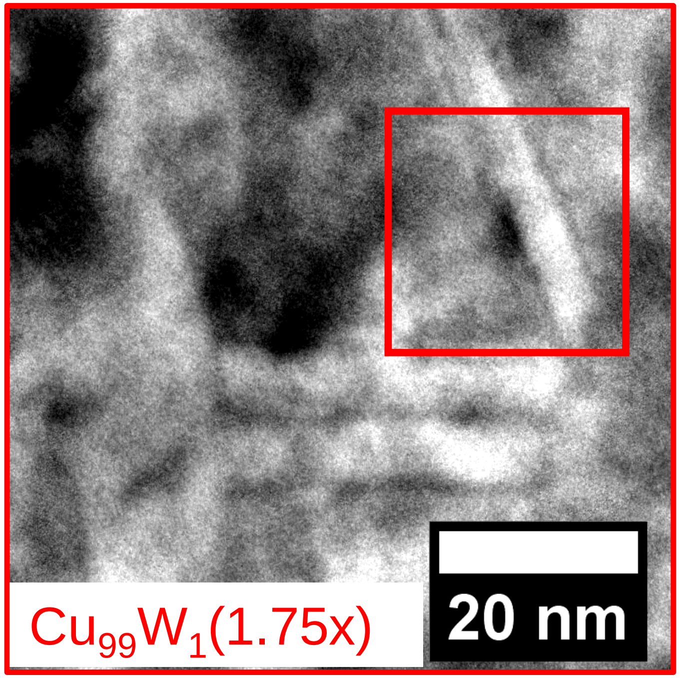

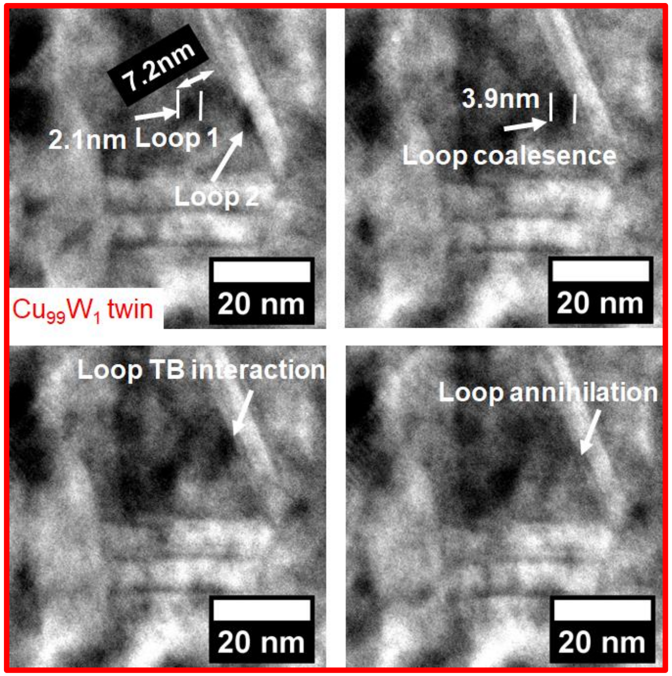

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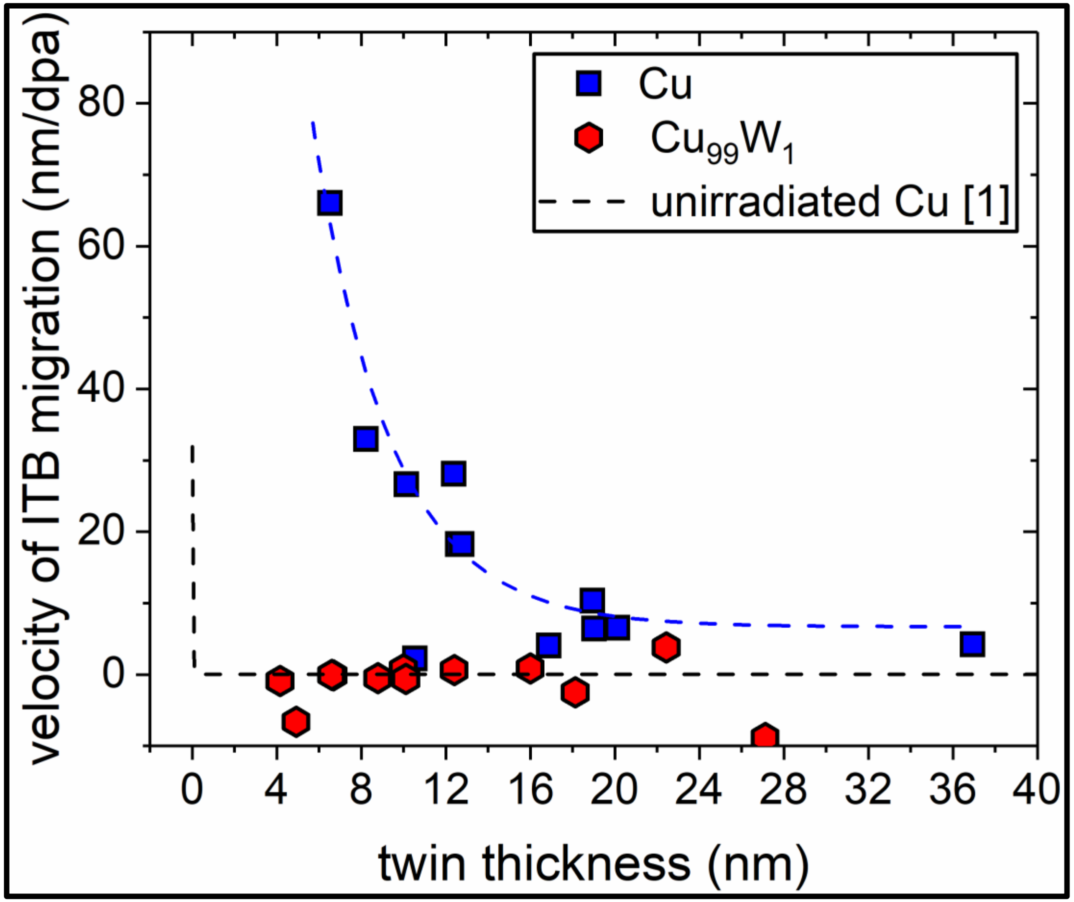
Detwinning rate of $Cu_{99}W_1$ is closer to that of unirradiated pure Cu at room temperature



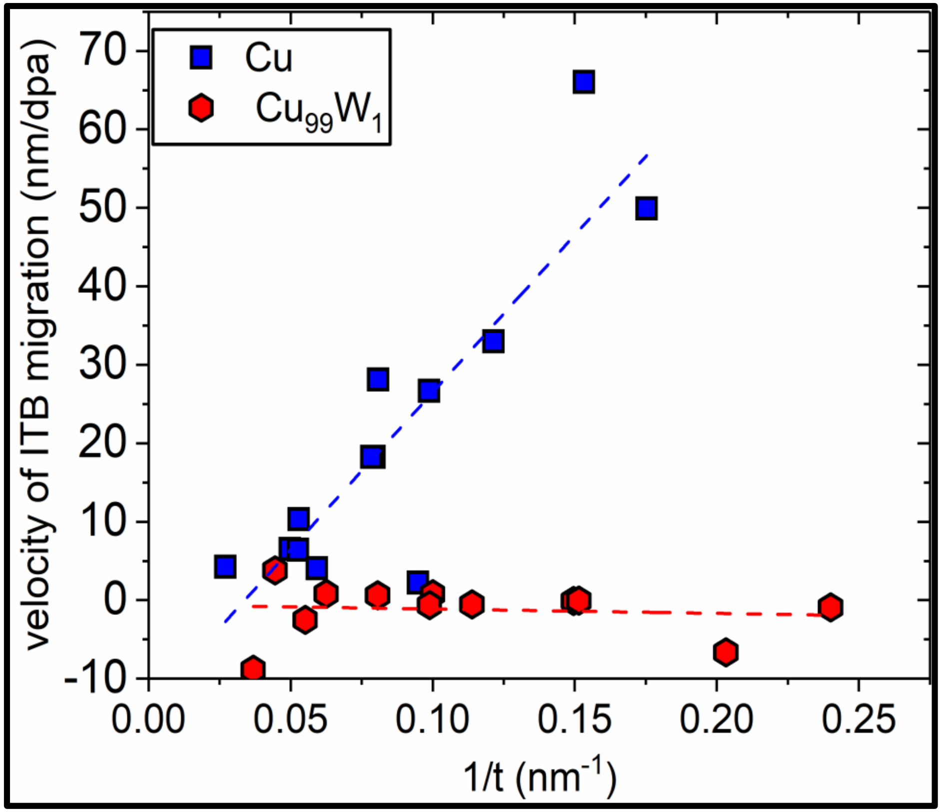
- →ITB migration rate for $Cu_{99}W_1$ is a result of change in grain dimension.
- →Certain twins show an increase in length in contrast to pure Cu.
- →Activation energy for ITB migration increases with defect concentration

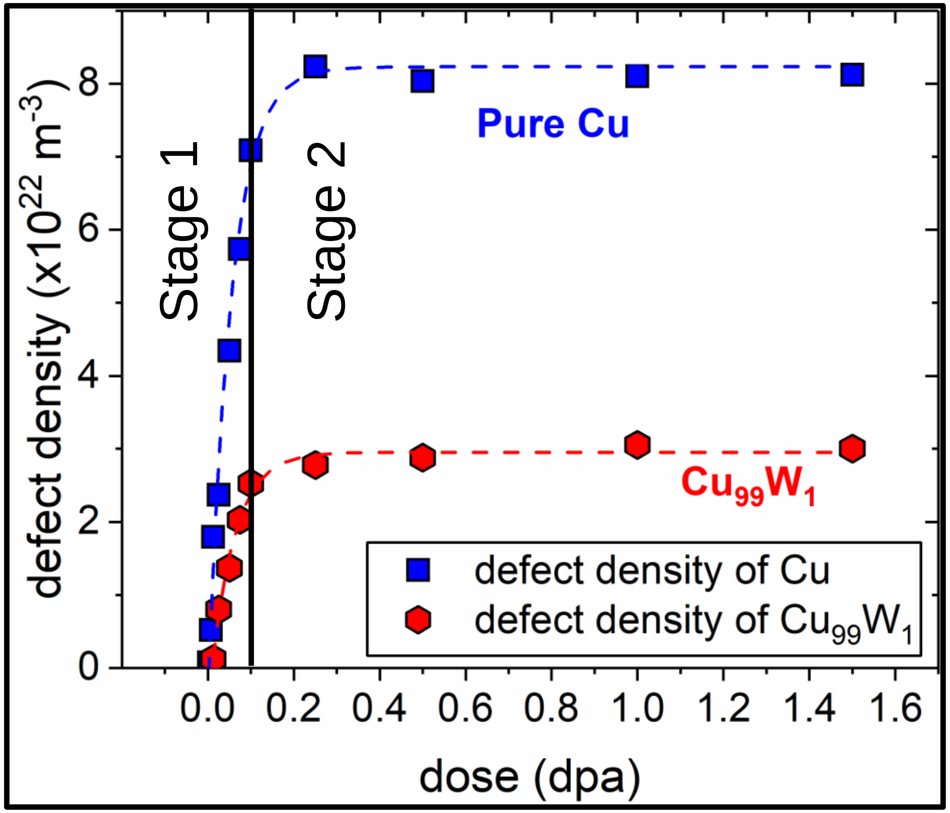
$$V = \frac{2\gamma}{t} M_0 e^{\left(-\frac{\Delta Q}{kT}\right)} \rightarrow \text{Velocity of Twin Migration}$$

$$\Delta Q = \Delta Q_0 \left(1 - \frac{\tau}{\tau^c}\right)^{\alpha} \rightarrow \text{Activation energy for Migration}$$

[1] L. Xu et. al. J. Appl. Phys. 104 (2008)

Defect density is significantly lower in $Cu_{99}W_1$ → greater activation energy and greater critical shear stress





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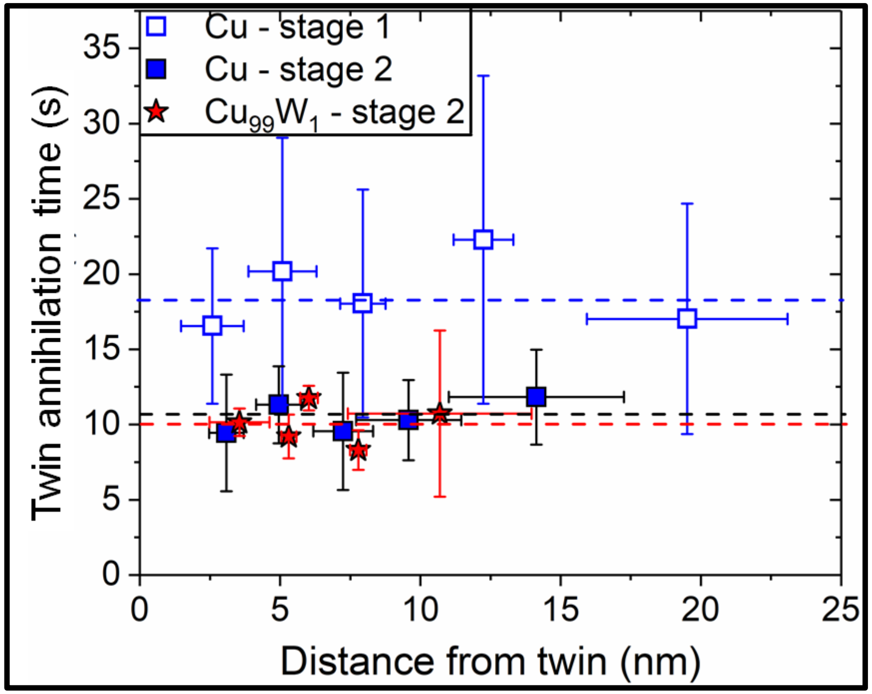
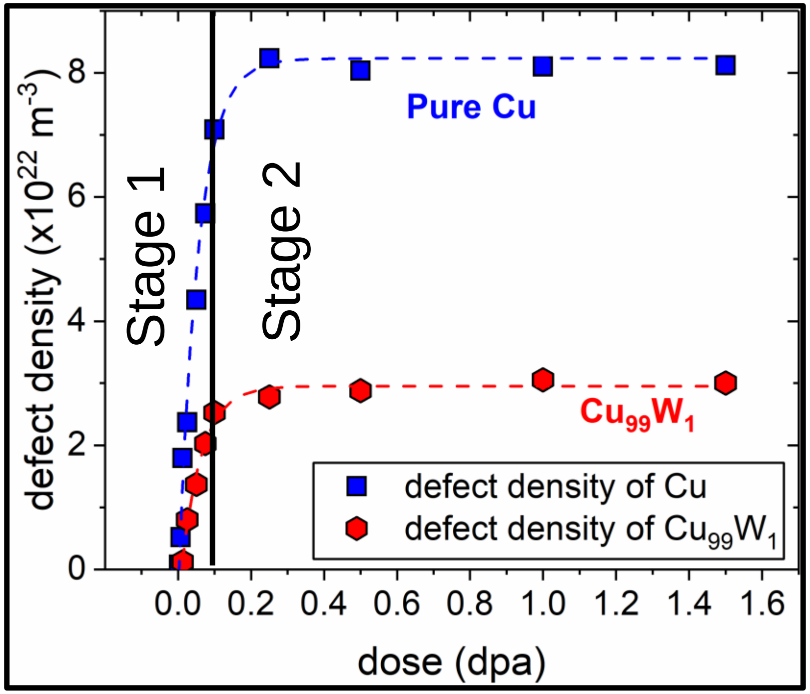
γ → excess CTB energy (24mJ/m^2)
t → twin thickness/twin spacing
M_0 → Defect mobility

Q_{cu} → ~0.34eV/atom
Q_{Cu99W1} >> Q_{Cu}

Defect annihilation time at twin boundaries is similar for both Cu and $Cu_{99}W_1$ and independent of distance from twin.

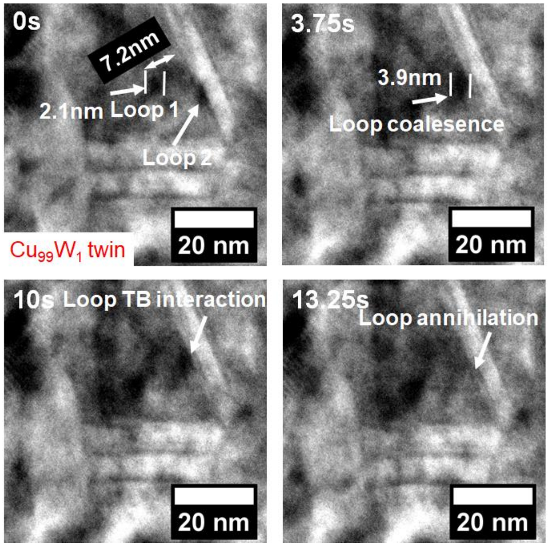


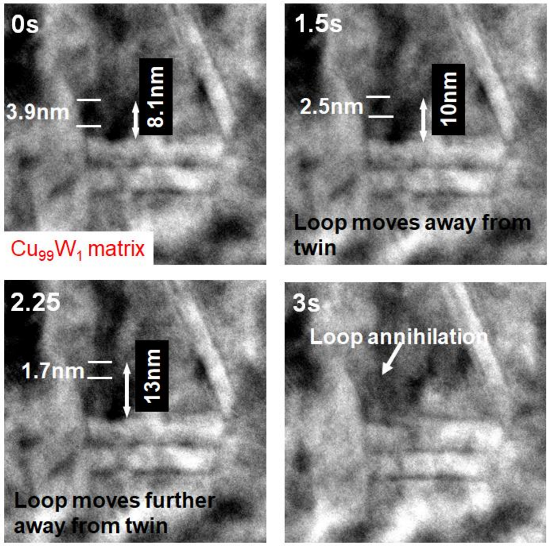


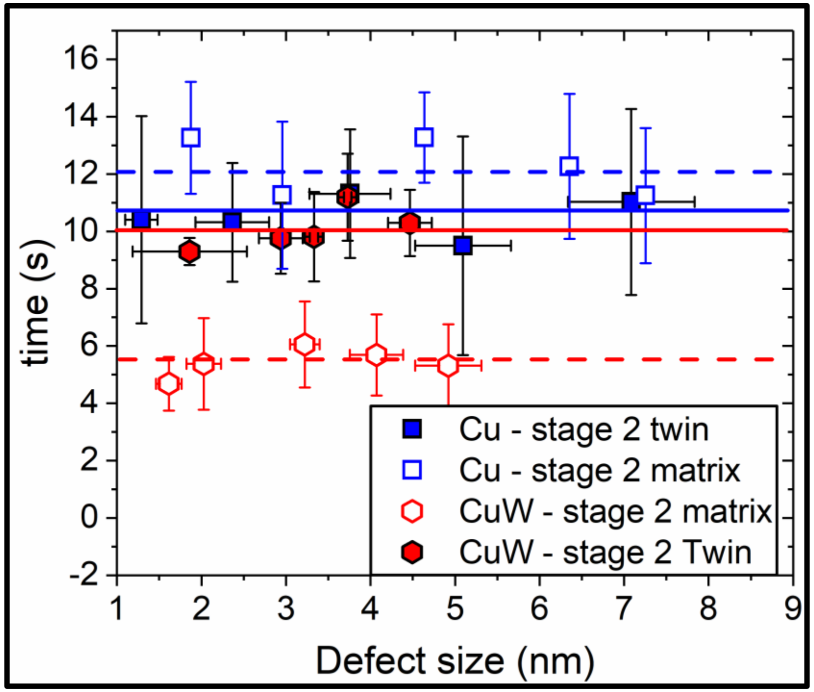
Greater defect density in Cu at higher doses (>0.1-0.2dpa) results in increased interactions of defects with CTBs and hence a shorter defect lifetime.

$Cu_{99}W_1$ has smaller twin annihilation time despite a smaller defect density as a result of smaller twin spacing ~3 times lower than that of Cu.

Defects with similar size distribution tend to annihilate faster in the matrix (non TB) compared to TB in $Cu_{99}W_1$



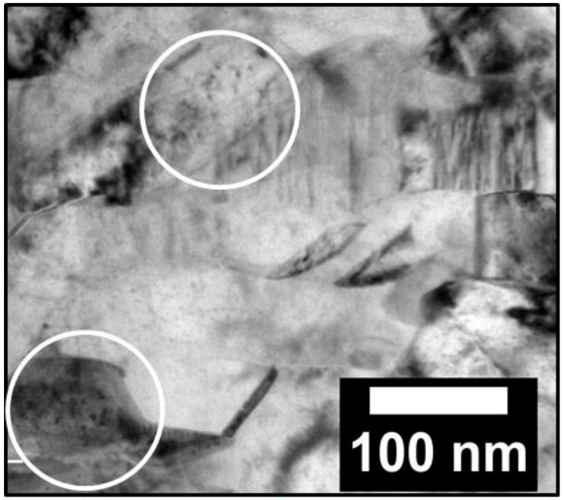


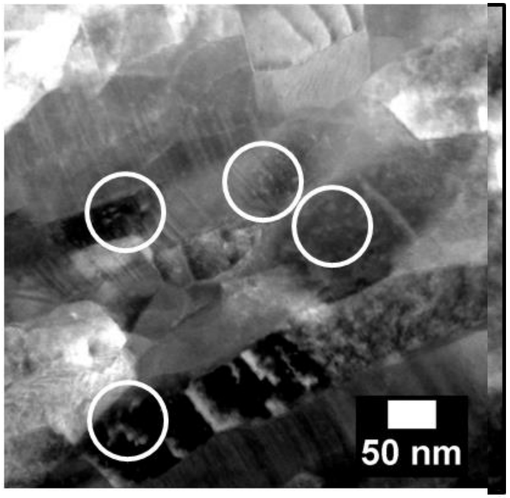


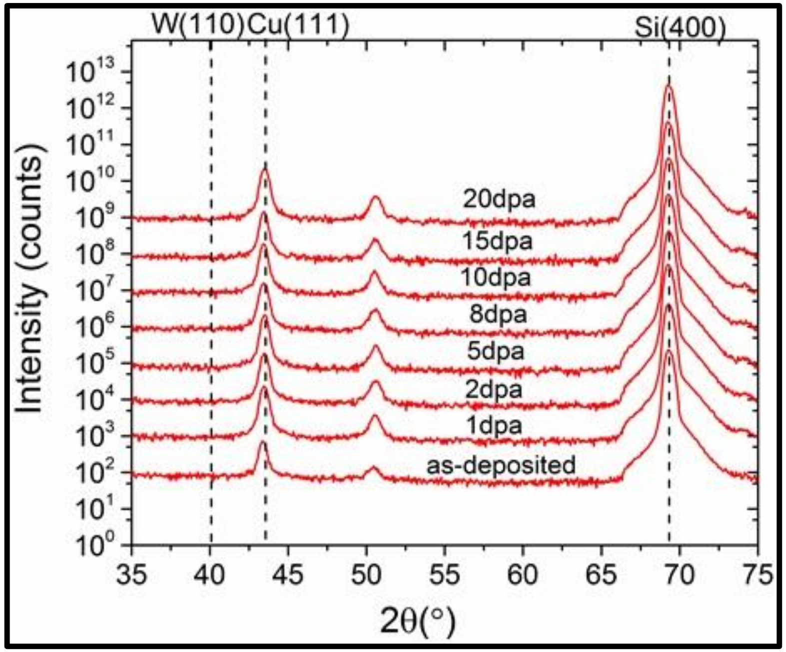
Sample	Defect lifetime (s)
Cu twin	**10.9**
Cu matrix	**13.2**
$Cu_{99}W_1$ twin	**10.1**
$Cu_{99}W_1$ matrix	**5.5**

Is the smaller defect lifetime in the matrix in $Cu_{99}W_1$ a result of defect absorption by isolated W clusters/precipitates that are formed during irradiation?

Ex-situ TEM and HAADF-STEM imaging clearly shows the presence of W precipitates in after irradiation in $Cu_{99}W_1$







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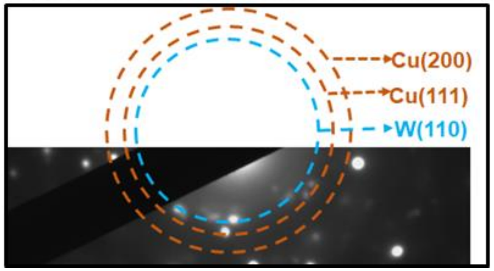


- → Steady state irradiation induced W solubility in Cu at room temperature is ~0.7at%.
- → Average particle size is ~2.7nm, average solute spacing is ~0.617nm.
- → Average precipitate spacing *(l)* calculated using $l = r_p\left[\left(\frac{1}{2f}\right)^{1/3} - 1\right]$ is ~10nm
- → Particle concentration c_p → $3.1\text{x}10^{-6}$ for $Cu_{99}W_1$

Twin boundaries and W precipitates act as competing sinks for radiation induced point defects

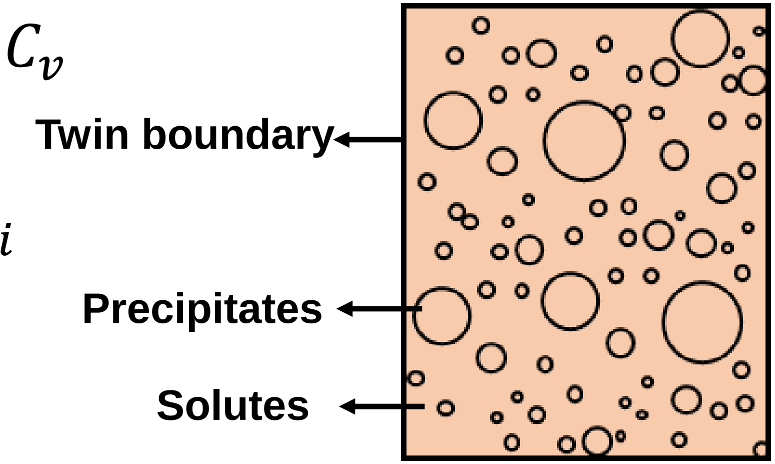
$$\frac{\partial C_v}{\partial t} = K_0 - K_{iv}C_iC_v - \eta K_{vs}C_vC_s + \Delta D_vC_v$$

$$\frac{\partial C_i}{\partial t} = K_0 - K_{iv}C_iC_v - \eta K_{is}C_iC_s + \Delta D_iC_i$$

Defect production rate

Recombination

Sink migration



$$\frac{\partial c_{i(v)}}{\partial t} = 0 = \xi_{FM}K_o - \frac{4\pi r_{iv}}{\Omega}(D_i + D_v)c_{i(v)}c_{v(i)} - \eta_p\frac{4\pi r_p}{\Omega}D_L^p c_L c_p - \eta_{twins}\frac{3\pi^2}{L_{twins}^2}D_L^{twins}c_L$$

Defect production

Recombination

precipitates

Twin boundary

η → sink efficiency, L → avg. twin spacing, c_i→ concentration of species i,
r_p → avg. precipitate size

In the sink limited regime recombination term is cancelled.
Equating third and fourth term gives the fraction annihilating at precipitates and twin boundaries.

R. Sizmann J. Nucl. Mater. 70 (1968).

W precipitates trap more defects compared to twin boundaries if we assume $\eta_p = \eta_{twins}$

$$\frac{\partial c_{i(v)}}{\partial t} = 0 = \xi_{FM} K_o - \cancelto{0}{\frac{4\pi r_{iv}}{\Omega}(D_L) D_L^2} - \underbrace{\eta_p \frac{4\pi}{\Omega}(D_L^p + D_L^s) c_L (r_p c_p + r_s c_s)}_{(\zeta_p)} - \underbrace{\eta_{twins} \frac{3\pi^2}{L_{twins}^2} D_L^{twins} c_L}_{(\zeta_{TB})}$$

L=12.96nm, c_p=3.1x10^{-6}, r_p =2.7nm, r_s =0.21nm, c_s=8.1x10^{-3}, Ω=1.582x10^{-2}

→ Diffusivity of defect loops prior to annihilation in matrix is $D_L^p + D_L^s = {(l_p^2 + l_s^2)}/{t_{matrix}}$

→ Diffusivity of defect loops prior to annihilation in TBs is $D_L^{twins} = {(L/2)^2}/{t_{twins}}$

→ Ratio (R) of defects annihilating at matrix/precipitate boundary to defects annihilating at twin boundaries R= $(\zeta_p)/(\zeta_{GB})$.

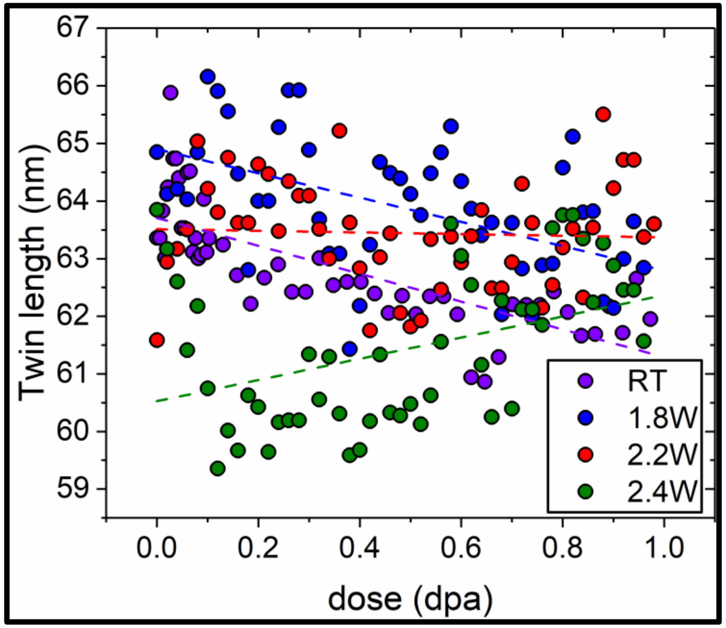
$D_L^p + D_L^s = {(l_p^2 + l_s^2)}/{t_{matrix}} = (10^2 + 0.617^2)/5.5$

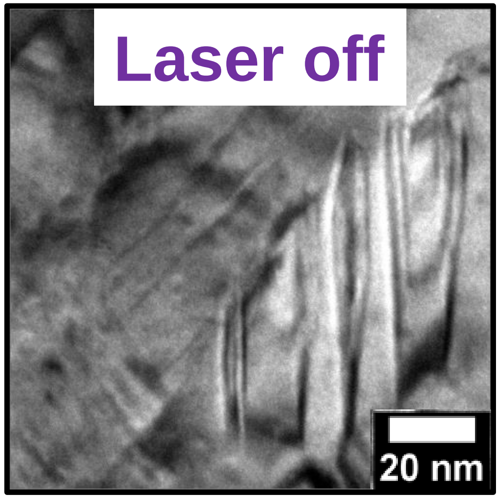
$D_L^{twins} = {(L/2)^2}/{t_{twins}} = (12.96/2)^2/10.1$

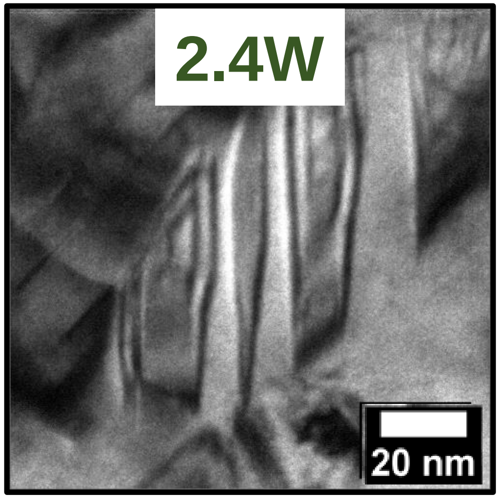
$$\boxed{R = (\zeta_p)/(\zeta_{TB}) = 33.89}$$

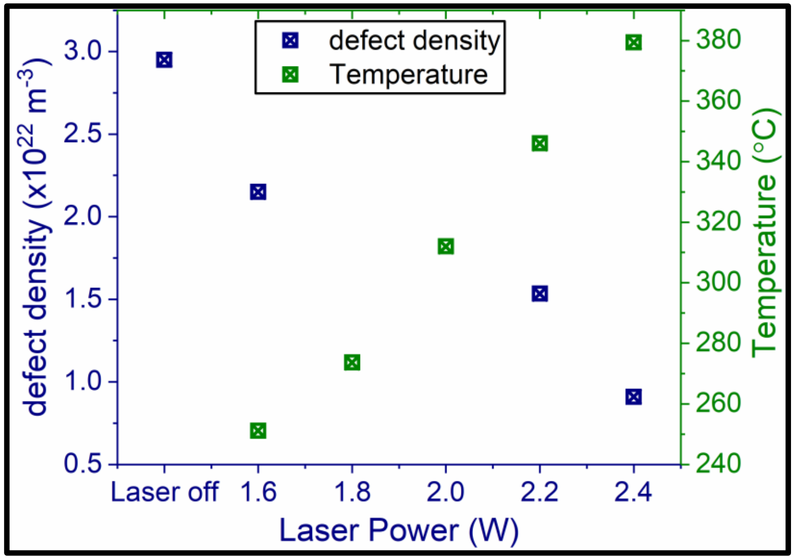
R. Sizmann J. Nucl. Mater. 70 (1968).

ITB nucleation and migration in $Cu_{99}W_1$ is insensitive to high temperature radiation









→Laser heating decreases the defect density and further increases the activation energy required for ITB migration.

→Small increase in twin length at 2.4W is a result of grain growth rather than ITB migration.

Summary and conclusions

→ The detwinning behavior of Cu and $Cu_{99}W_1$ was studied with the help of the I^3TEM available at Sandia National laboratories.

→ The detwinning rate of $Cu_{99}W_1$ is almost insignificant compared to that of pure Cu

→ The lower defect density results in a larger activation energy for detwinning in $Cu_{99}W_{1}$.

→ The average defect annihilation time at twins is the same for the both the samples but loops in the alloys annihilate faster in matrix owing to superior efficiency of W precipitates.