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The Remarkable Friction Behavior of Copper at Cryogenic Temperatures

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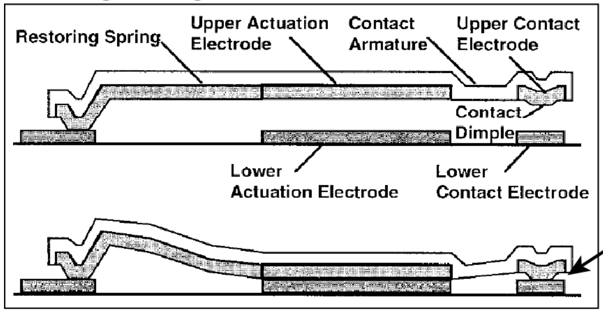
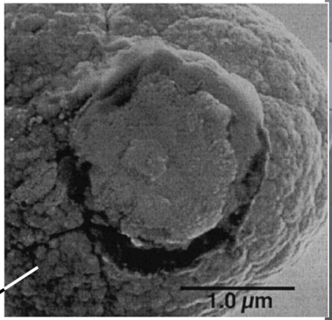
Metals are widely used tribological materials – particularly, electrical contacts

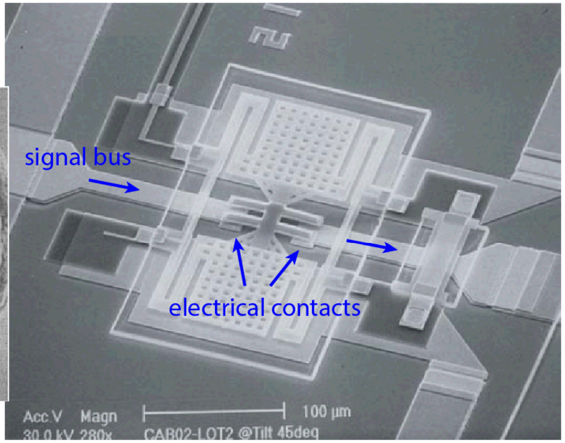
RF Micro Electromechanical Systems (MEMS)

switching GHz signals





Source: D. Hyman and M. Mehregany, IEEE Trans. & Pack. Tech. 22-3, 1999



Source: Rockwell Scientific metal-metal switch

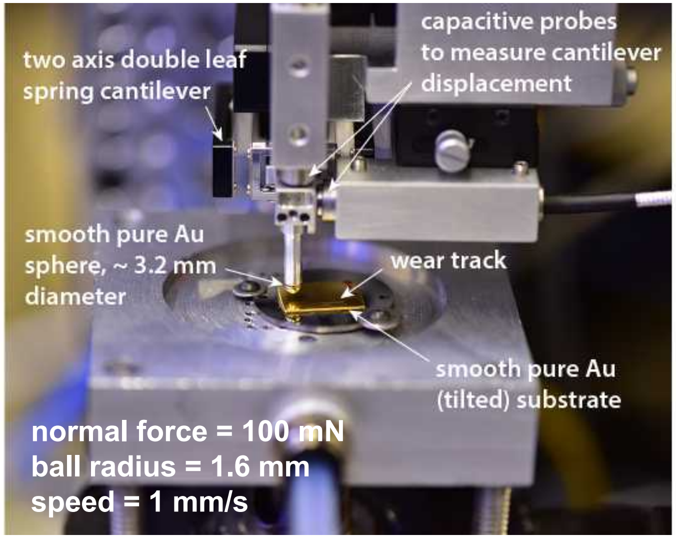
Electronics (e.g. PCB blade connectors):

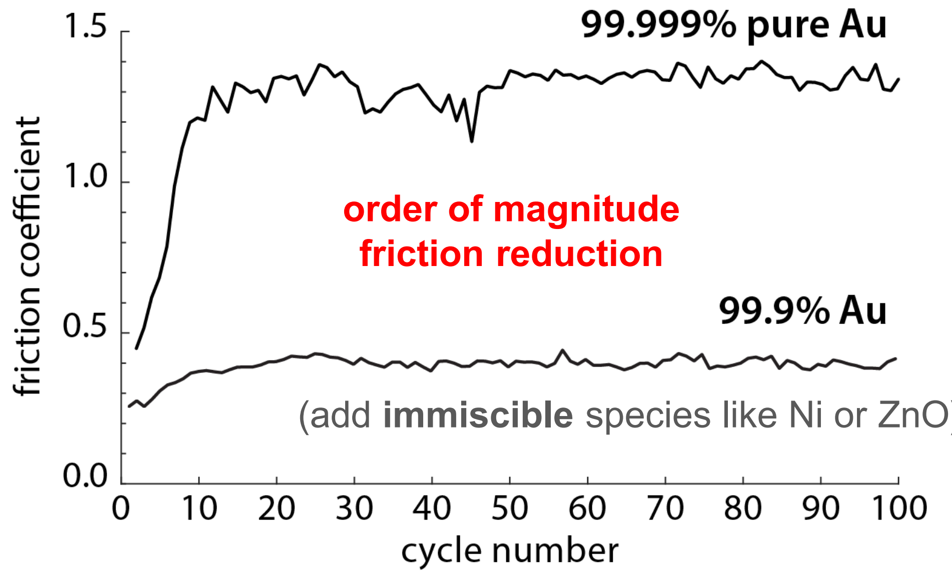
200 - 500 nm thick electroless hard gold

Aerospace and Energy

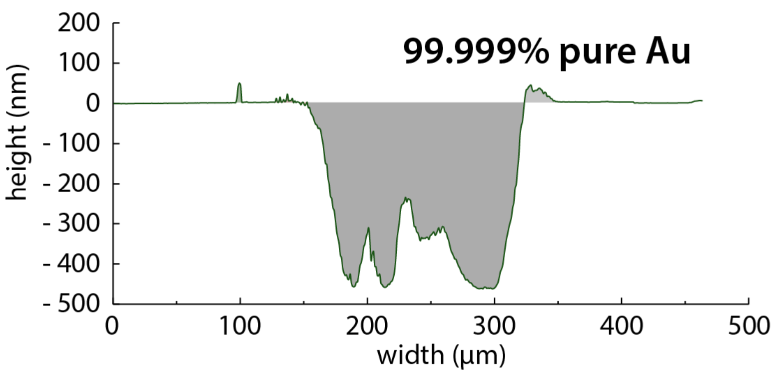
Source: Honeybee Robotics (http://www.honeybeerobotics.com/portfolio/rolling-contact-connector/)

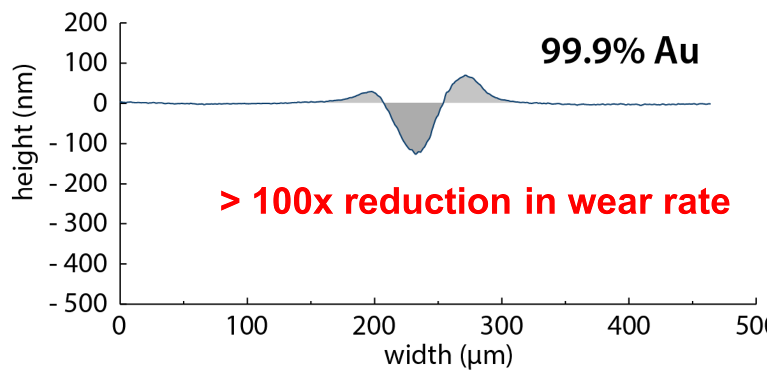
Examples of typical friction behavior of pure and alloyed (hard) Au films





Wear track cross-sections:



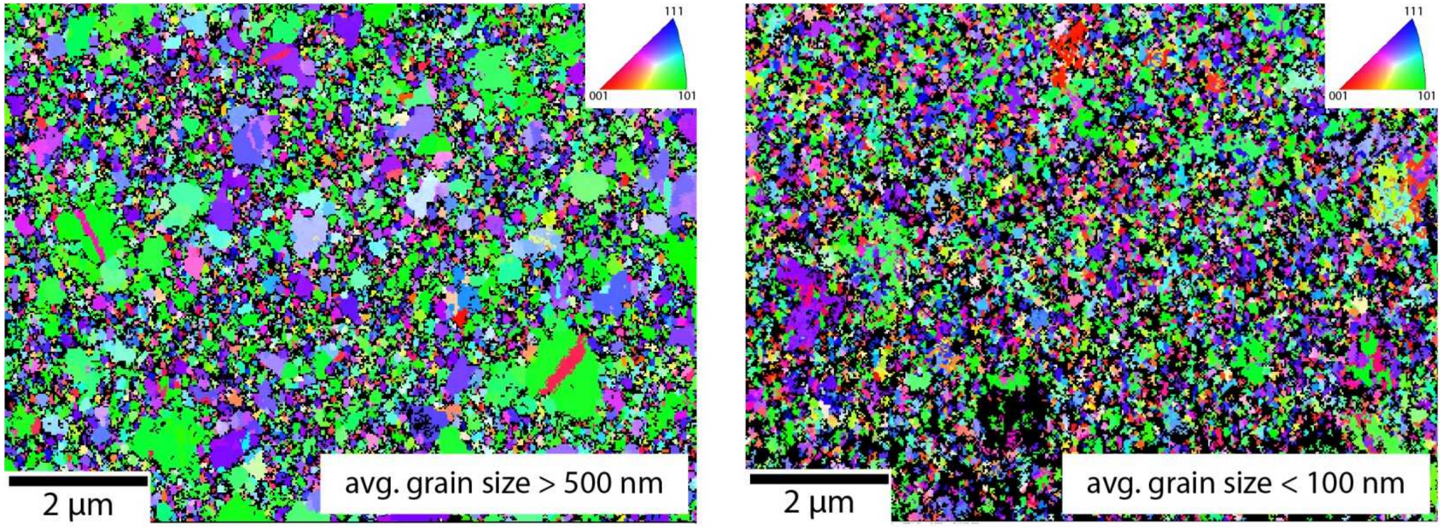


Same contact conditions, same environment, persistent result (> 10k cycles)
… so, *how?*

Alloying species can pin boundaries, reduce grain size (… and growth)

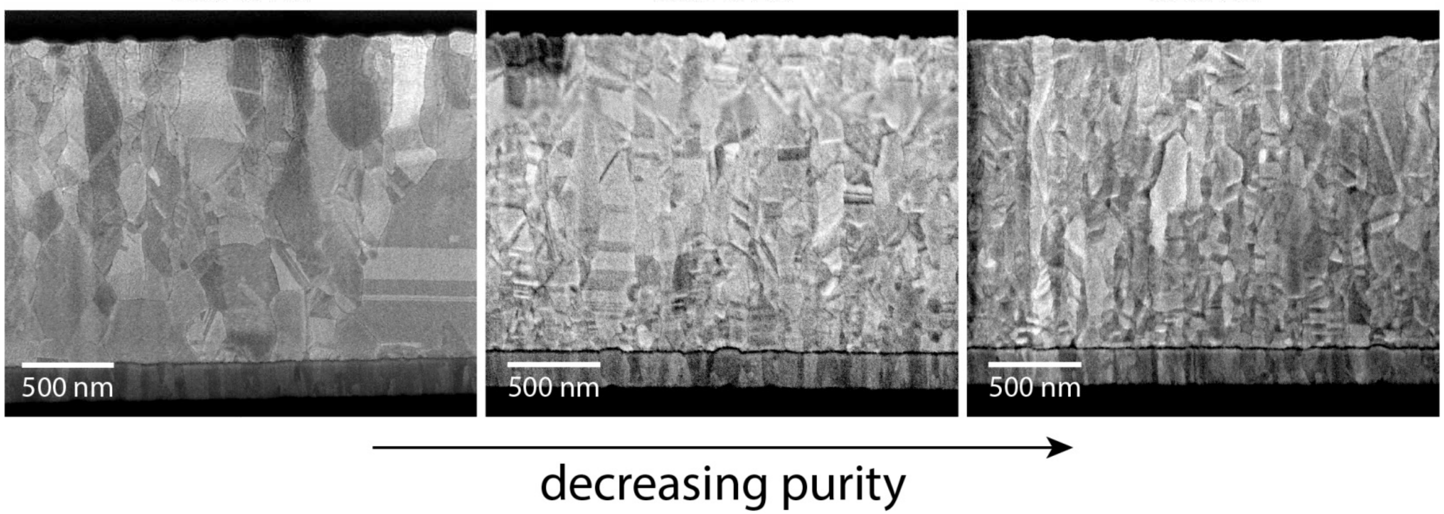
EBSD maps of film surfaces:

99.999% pure Au (film) | **99.9% hard Au film**

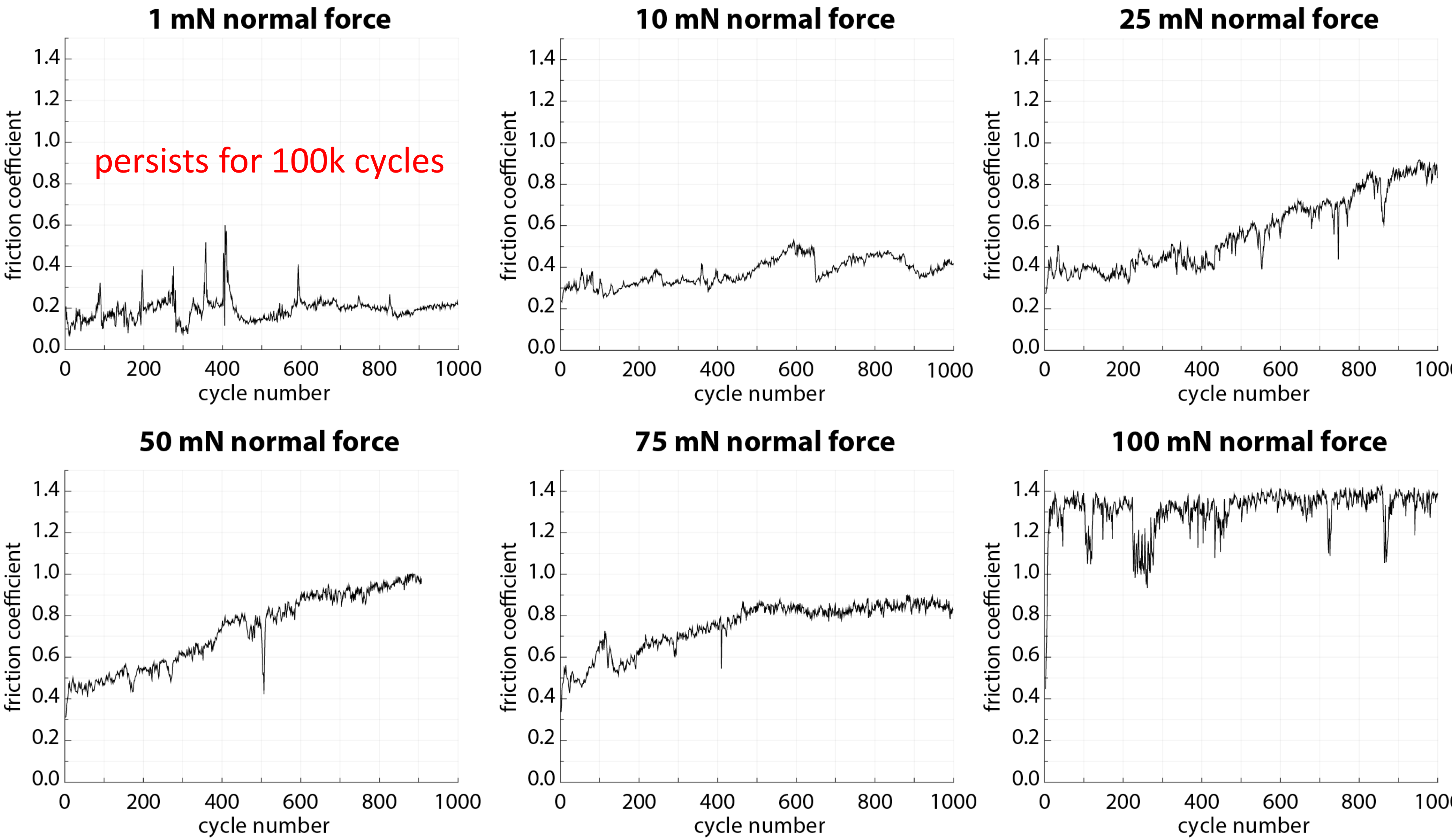


Film cross-sections (ion etched):

99.9% Au | **99.5% Au** | **99% Au**



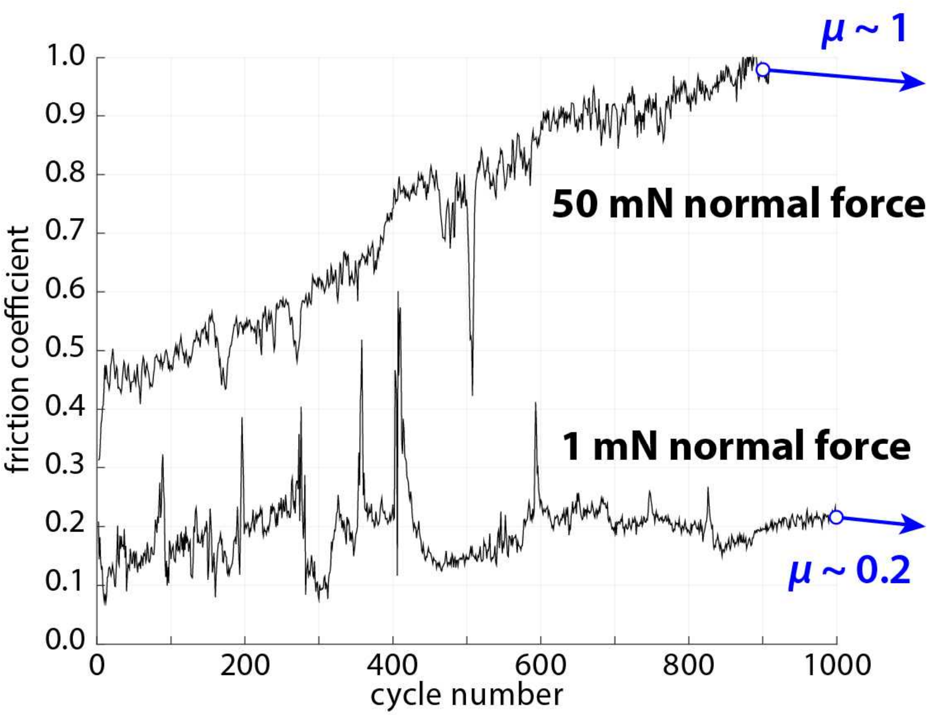
There are multiple friction regimes!



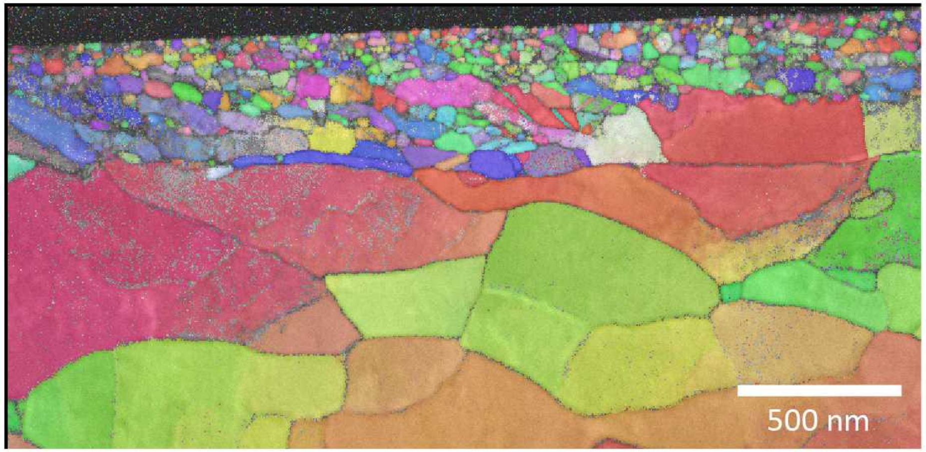
Pure Au in dry N_2

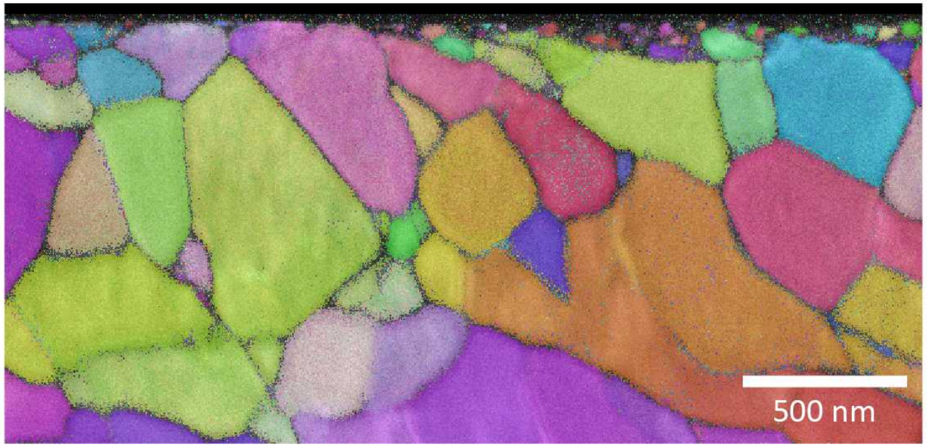
Electron diffraction of high and low friction wear tracks from Au-Au sliding contacts

Electron microscopy of focused ion beam prepared wear track cross-sections



Transmission Kikuchi Diffraction (TKD):
(transmission diffraction performed in an SEM)



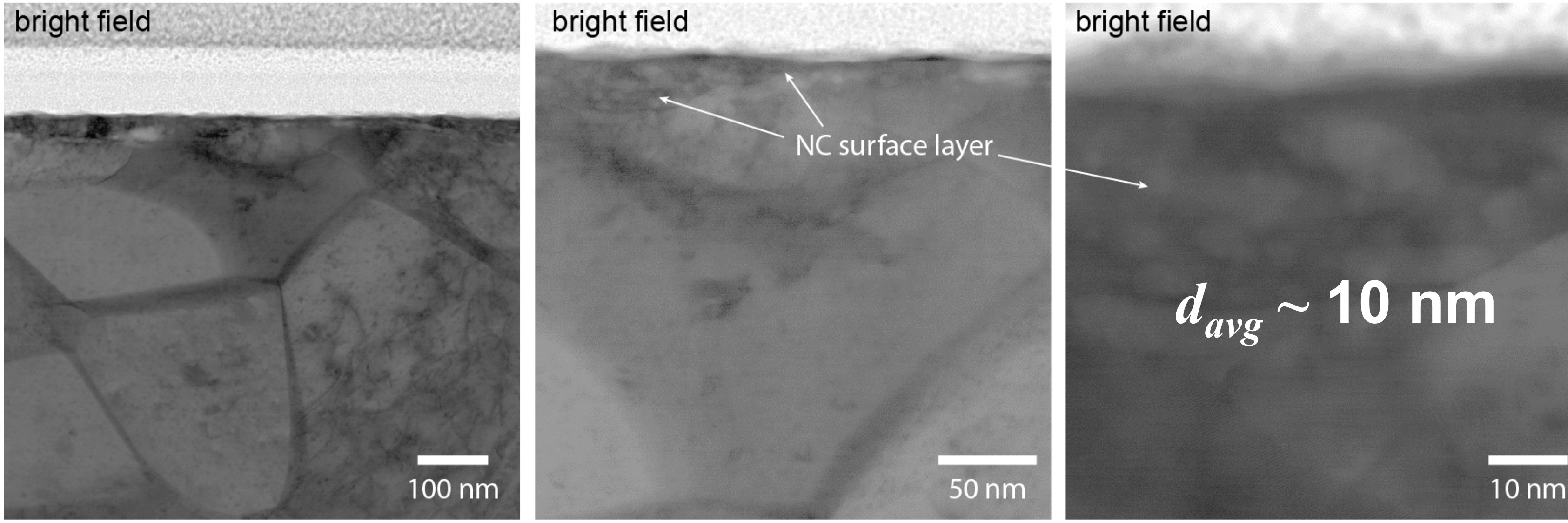


Again we see fine grain size in both cases… but the low friction case is smaller.

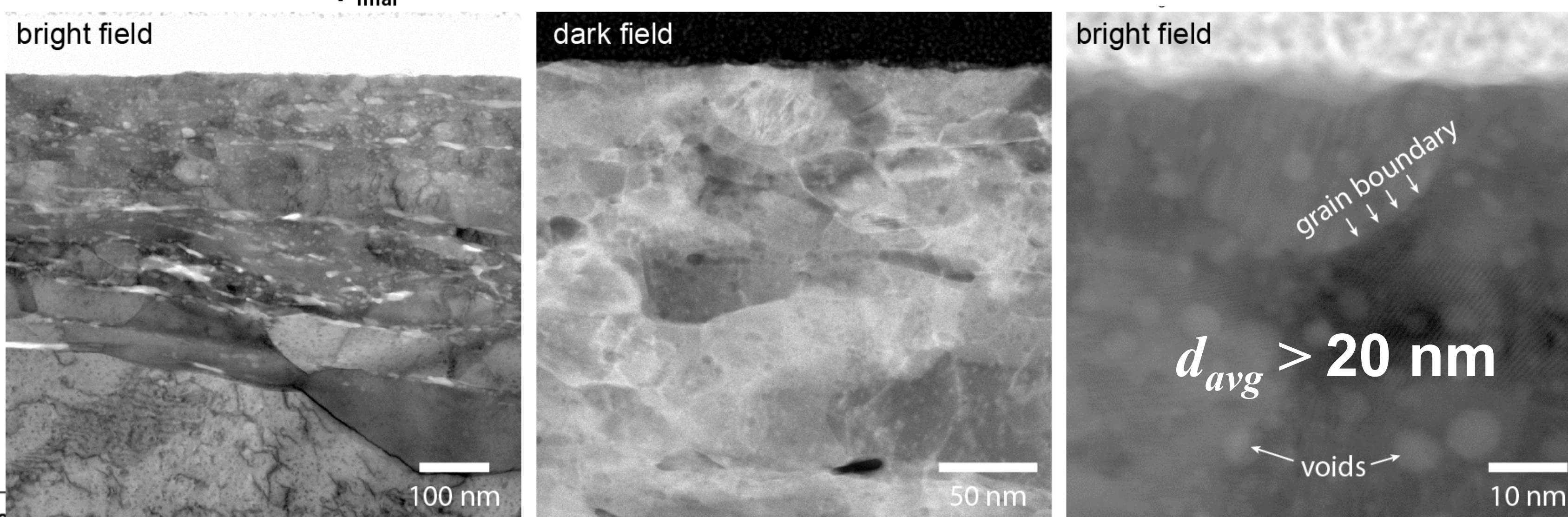
What is the connection between grain size and friction?

The difference in surface grain size between low/high friction is small

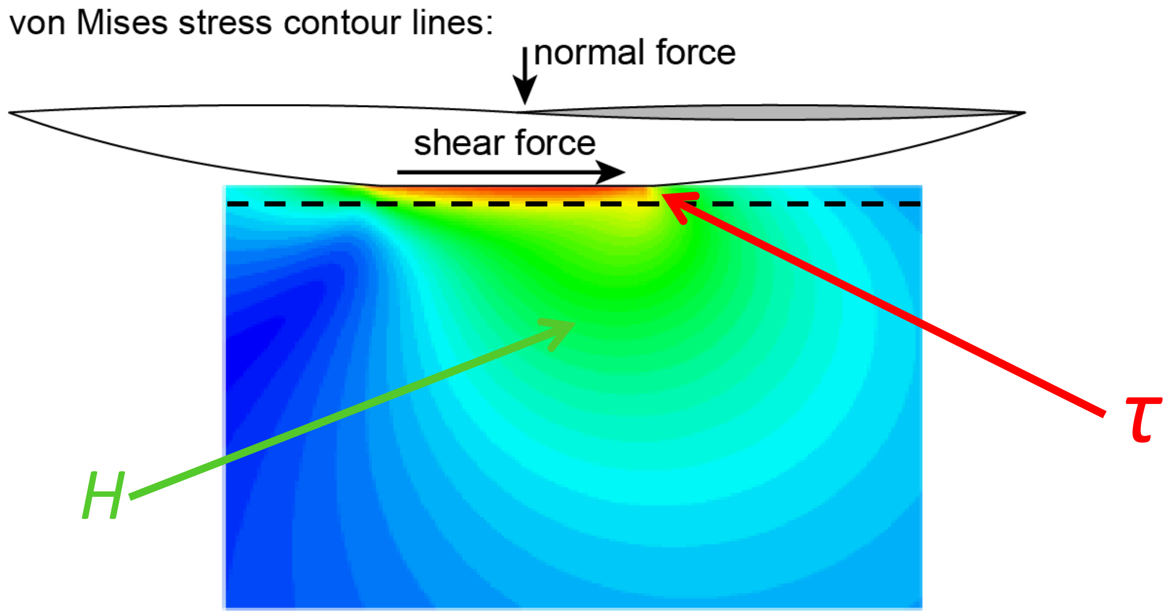
1 mN normal force, μ_{ss} ~ 0.2



50 mN normal force, μ_{final} ~ 1.0



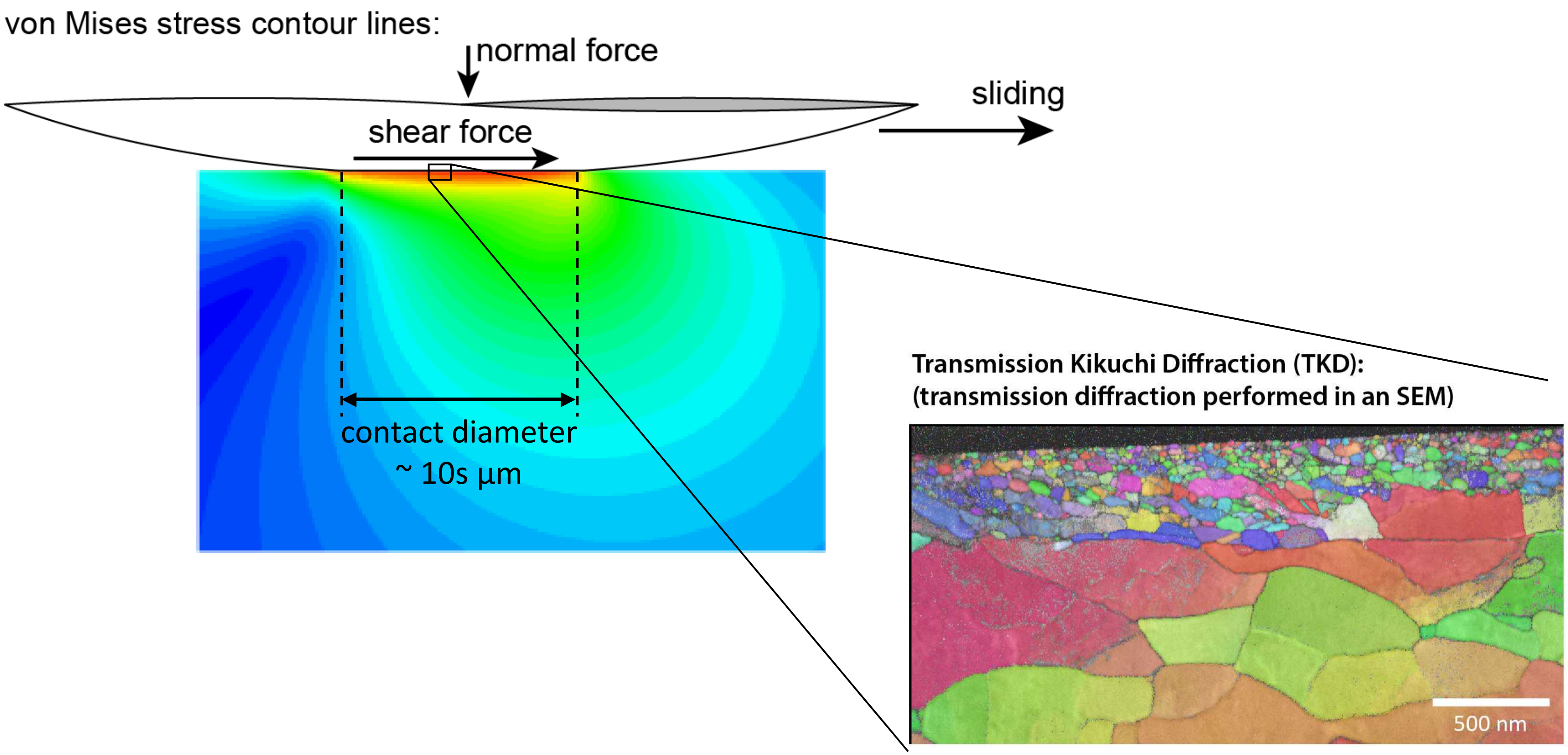
The basis of friction in metals



Bowden & Tabor: $\mu \cong \tau / H$

- τ: determined by *surface* grain size, gives μ
- H: determined by *bulk* grain size, gives contact area
- Surface evolves, bulk doesn't (much)
- TEM corroborates

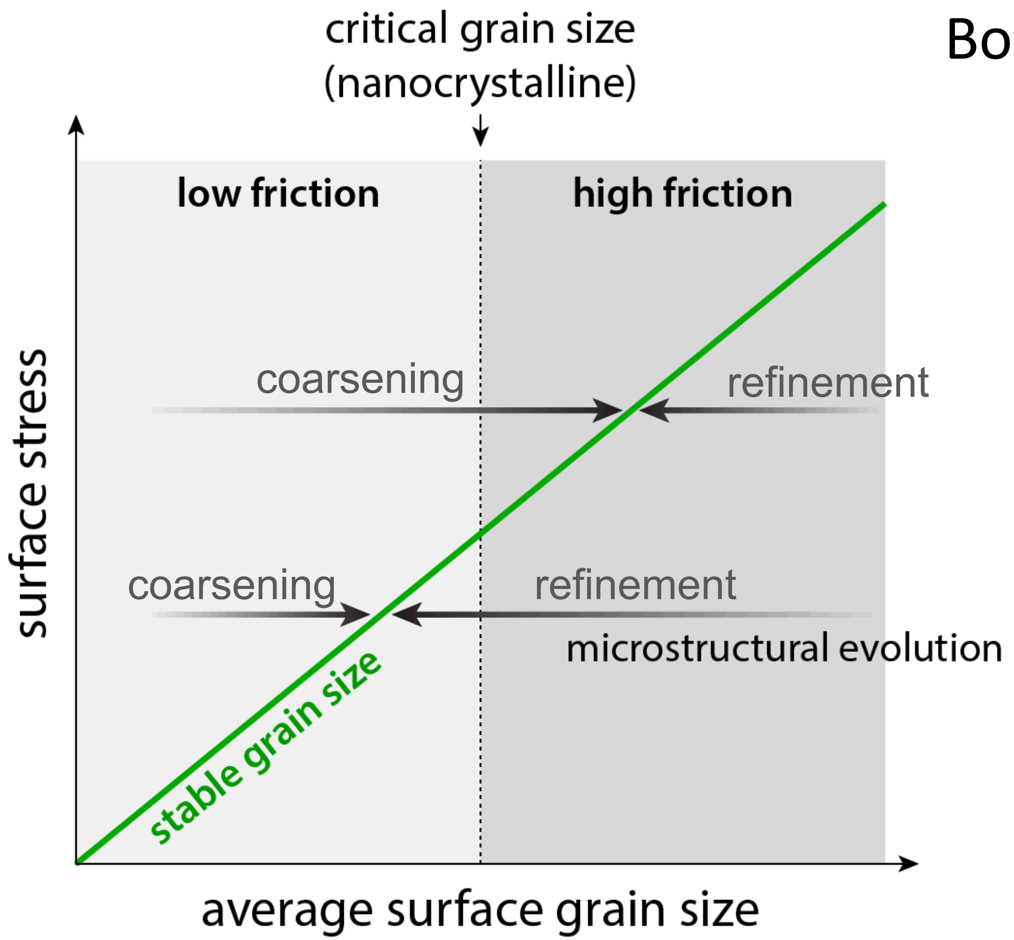
So how does interface strength impact friction coefficient so much?



depth of deformation layer is
an order of magnitude smaller!

This illustrates how interface shear strength (HIGHLY surface localized) and load bearing strength (bulk hardness) impact friction coefficient

We propose that there exists a stress-dependent steady-state (asymptotic) grain size



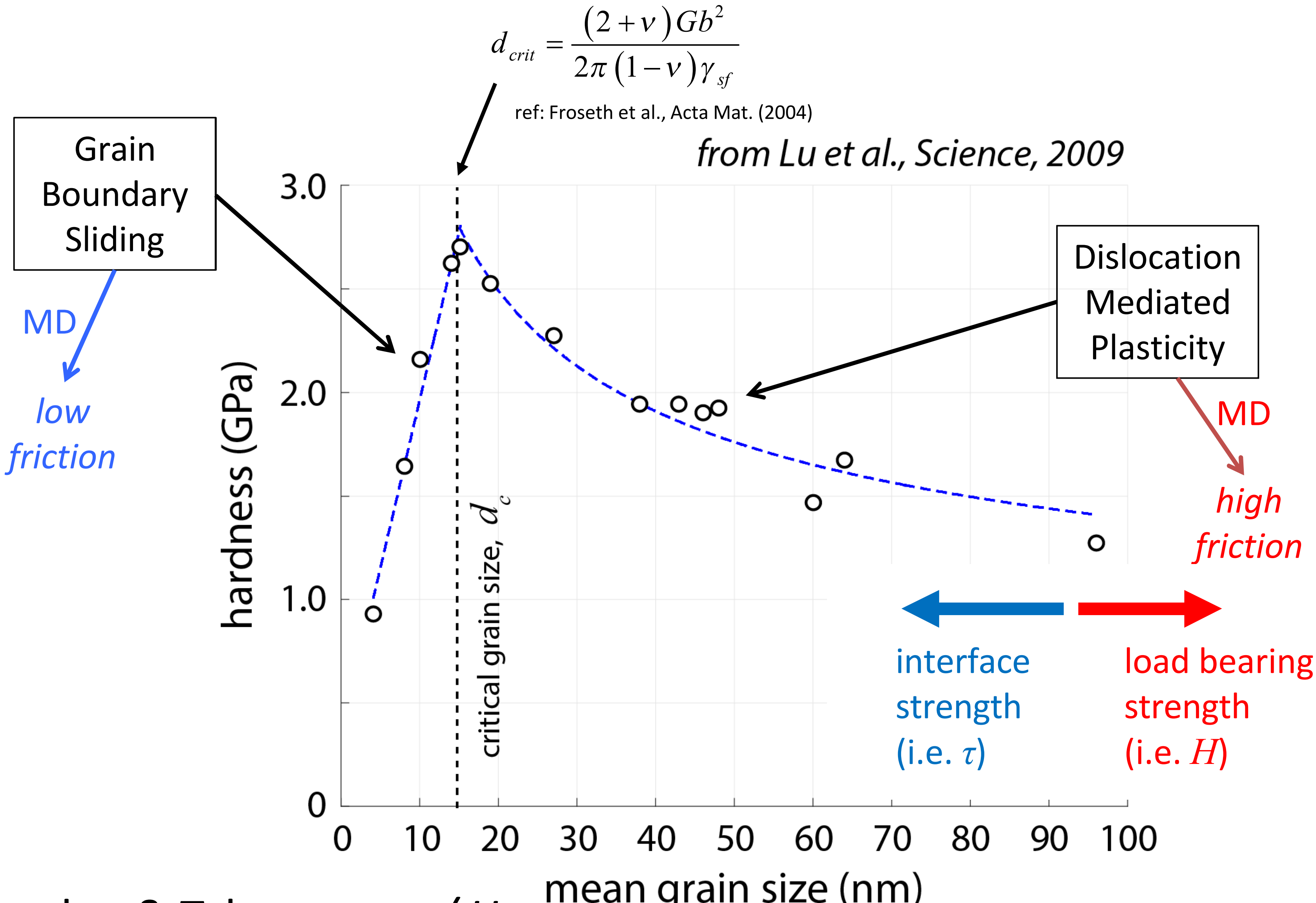
Bowden & Tabor: $\mu \cong \tau / H$

"All models are wrong, but some are useful."
--George Box

"It is better to be roughly right than precisely wrong."
--John Maynard Keynes

- Geologists have proposed this concept for rocks and ice cores (Derby et al., 1992)
- Was later extended to metals under severe plastic deformation (Pougis et al. in 2014)

How does grain size affect materials properties? Let's look at copper…

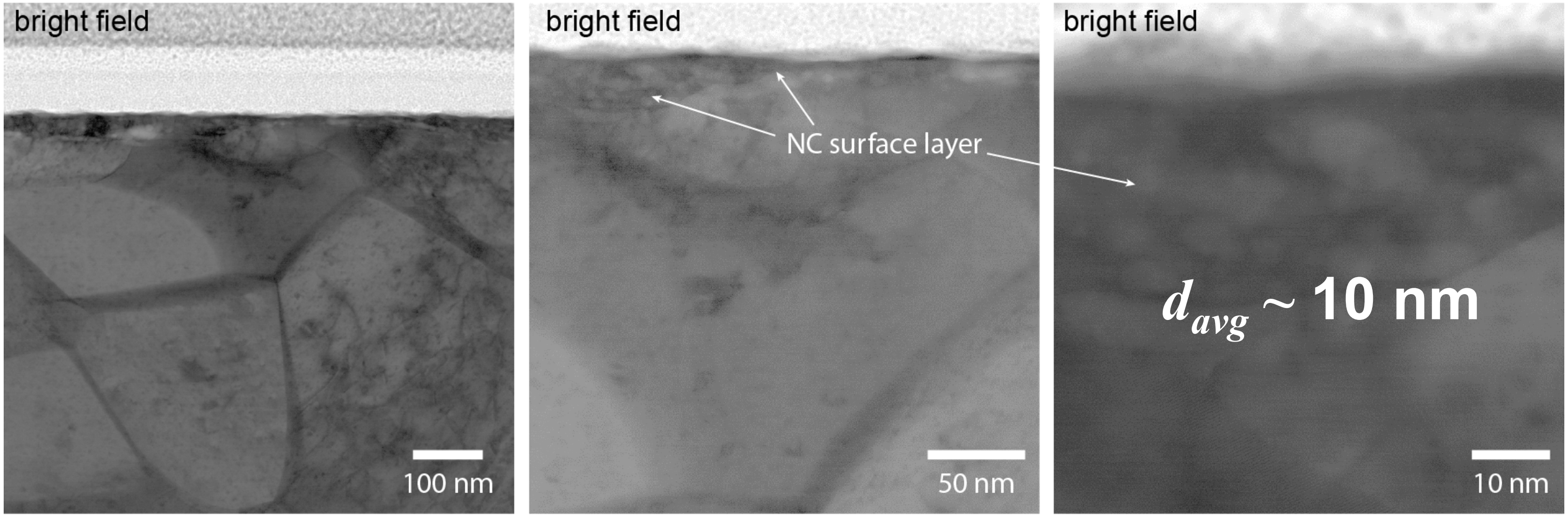


Bowden & Tabor: $\mu \cong \tau / H$

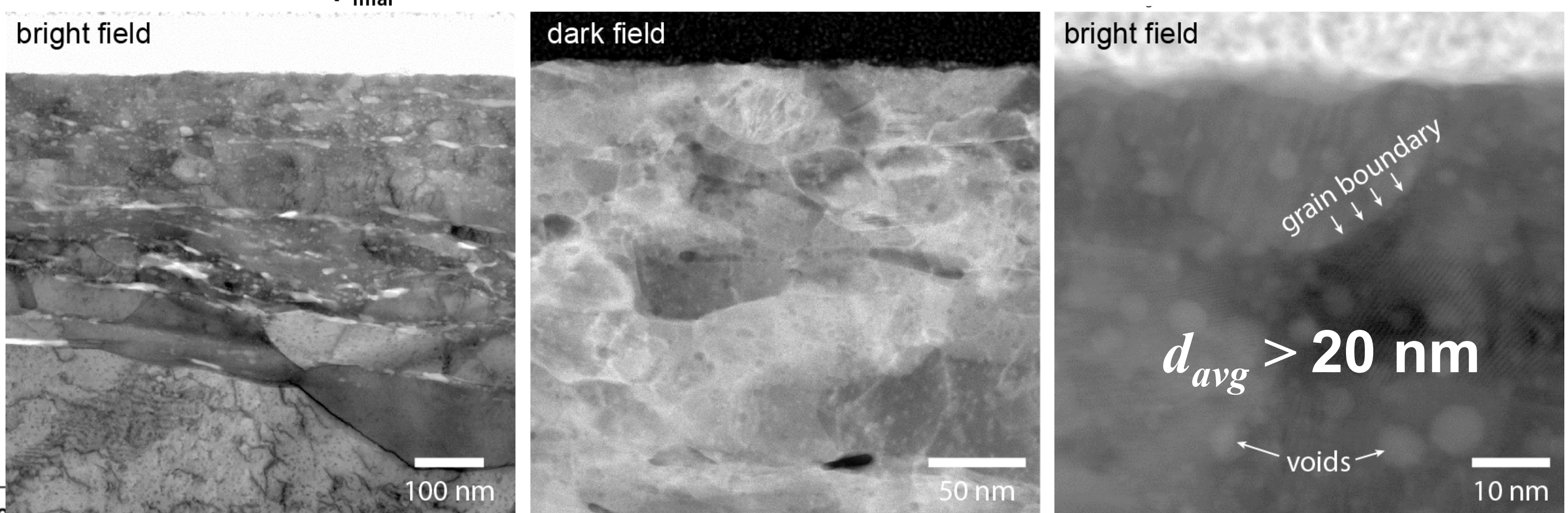
One last look at Au… critical grain size predicted from materials properties only!

critical grain size: $d_{crit,\,Au} = 17.4\ nm$

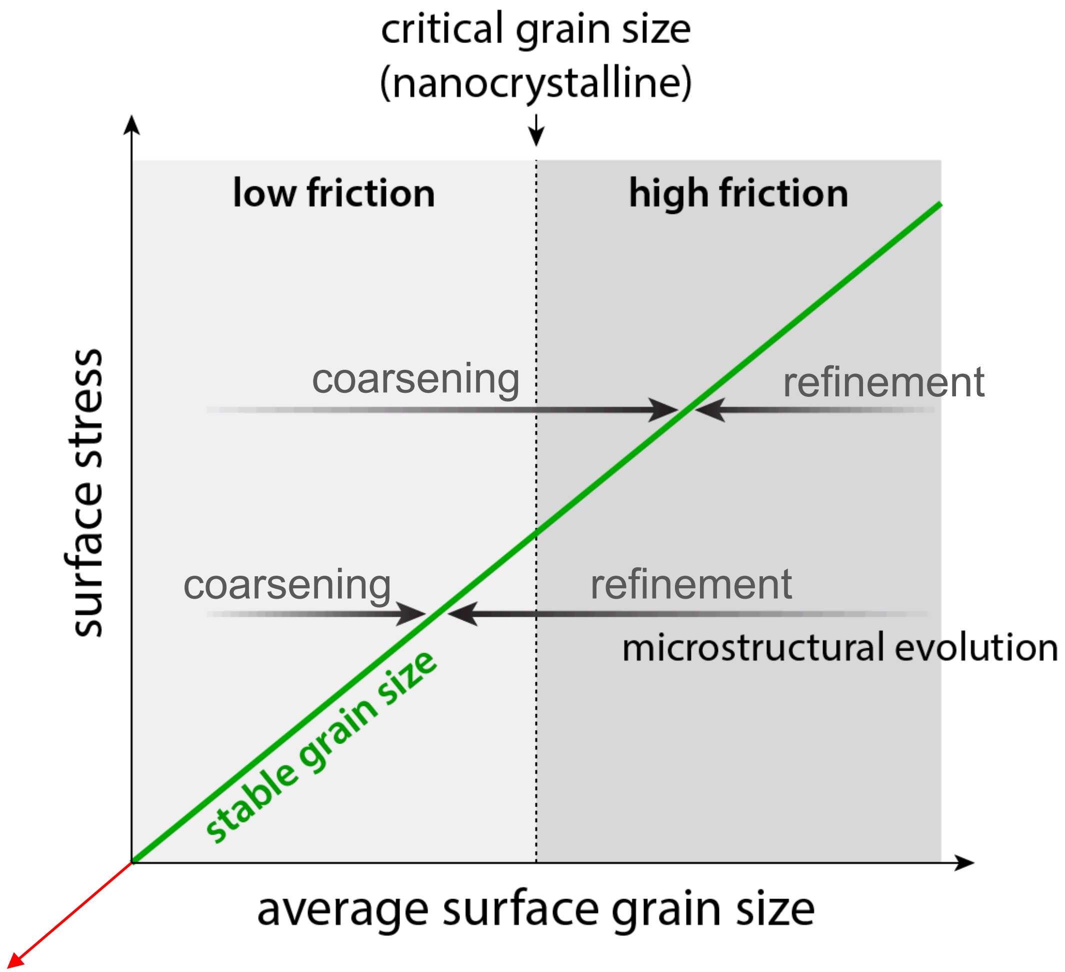
1 mN normal force, μ_{ss} ~ 0.2



50 mN normal force, μ_{final} ~ 1.0



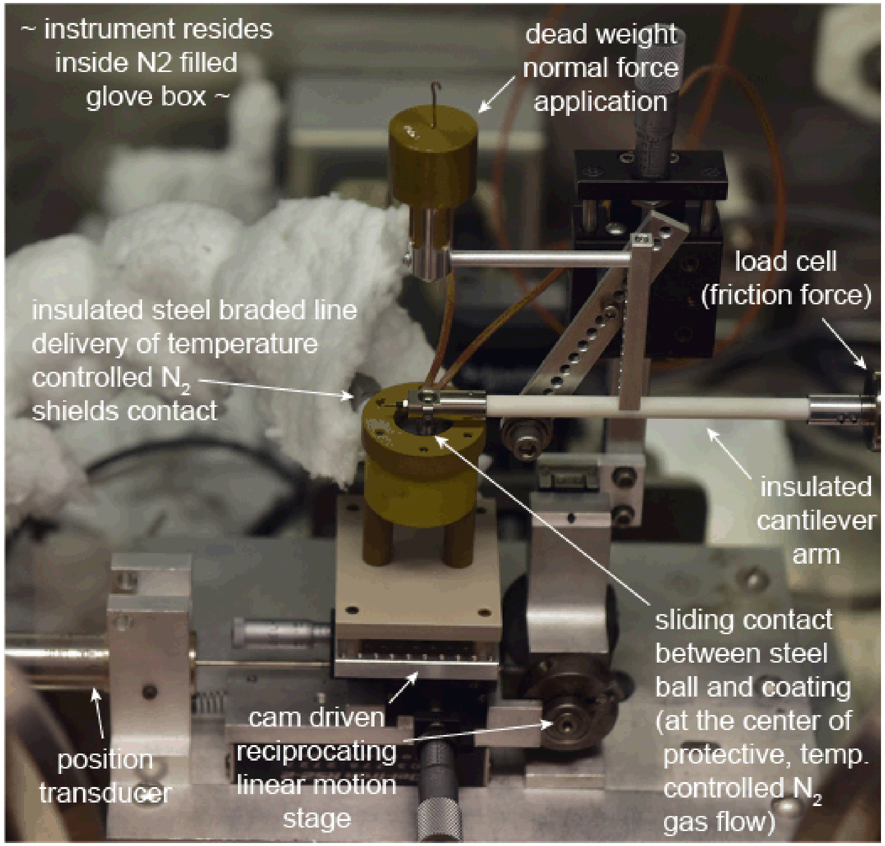
If stress-dependent grain growth kinetics matter, then temperature should work!

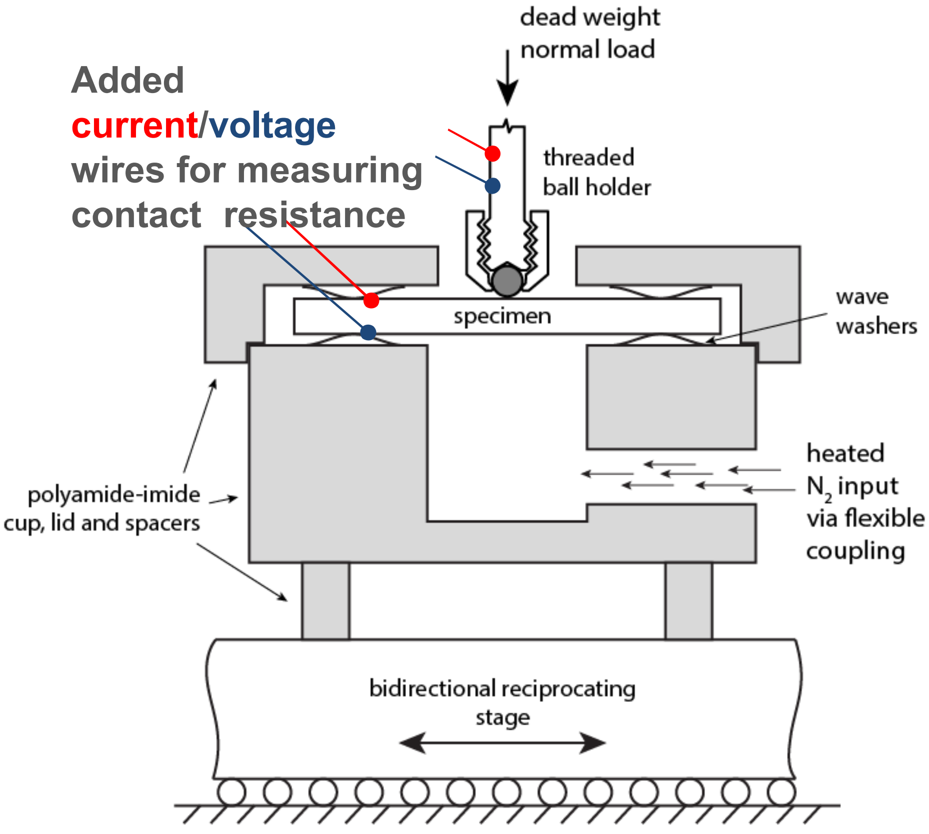


What about reducing grain growth kinetics by lowering the temperature?

Cryogenic experiments can help us verify

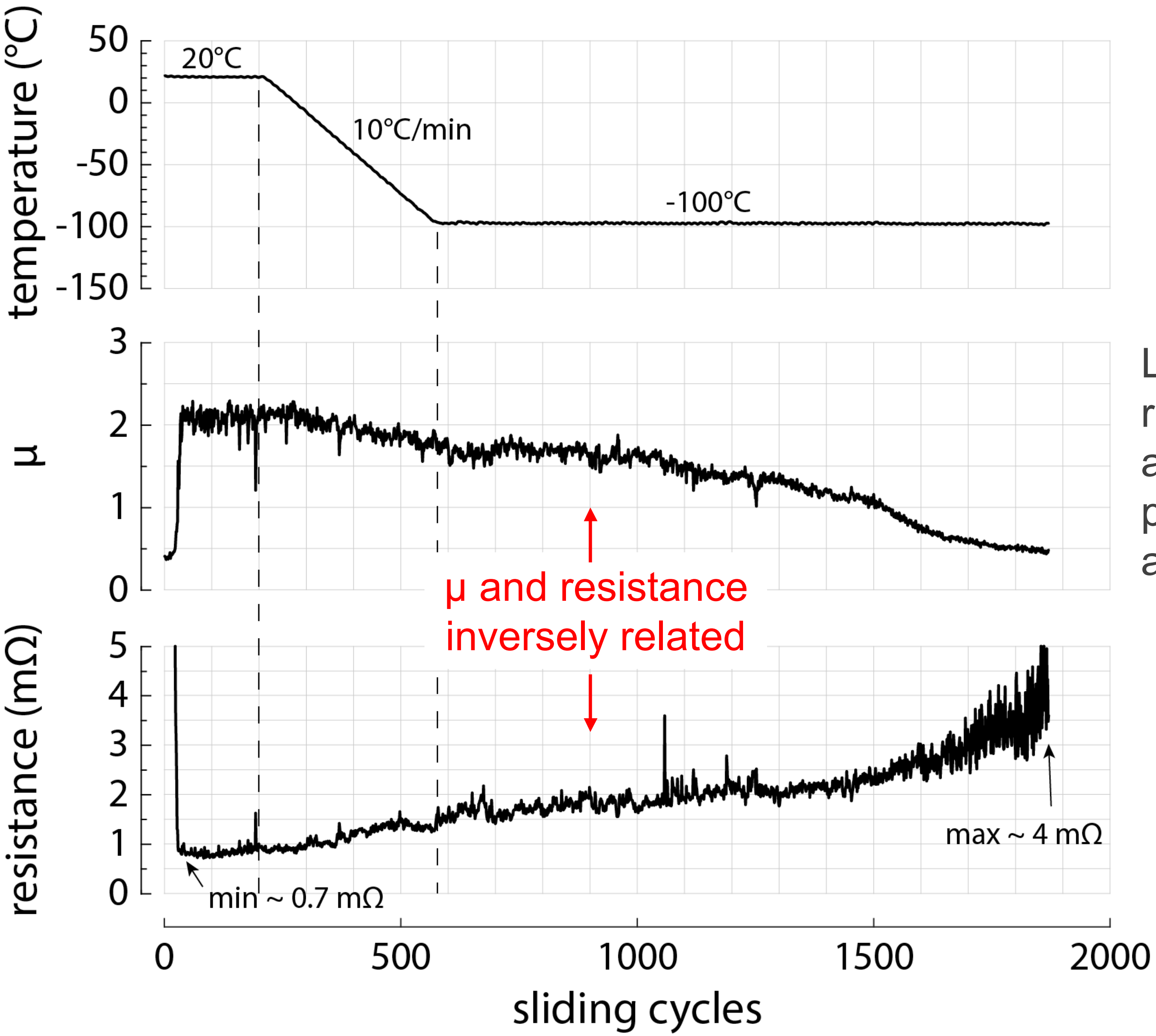
variable temperature tribometer (-190°C to +250°C) in inert gas environment (liq. N_2 input)





(< 10 ppm O_2 and < 10 ppm H_2O)

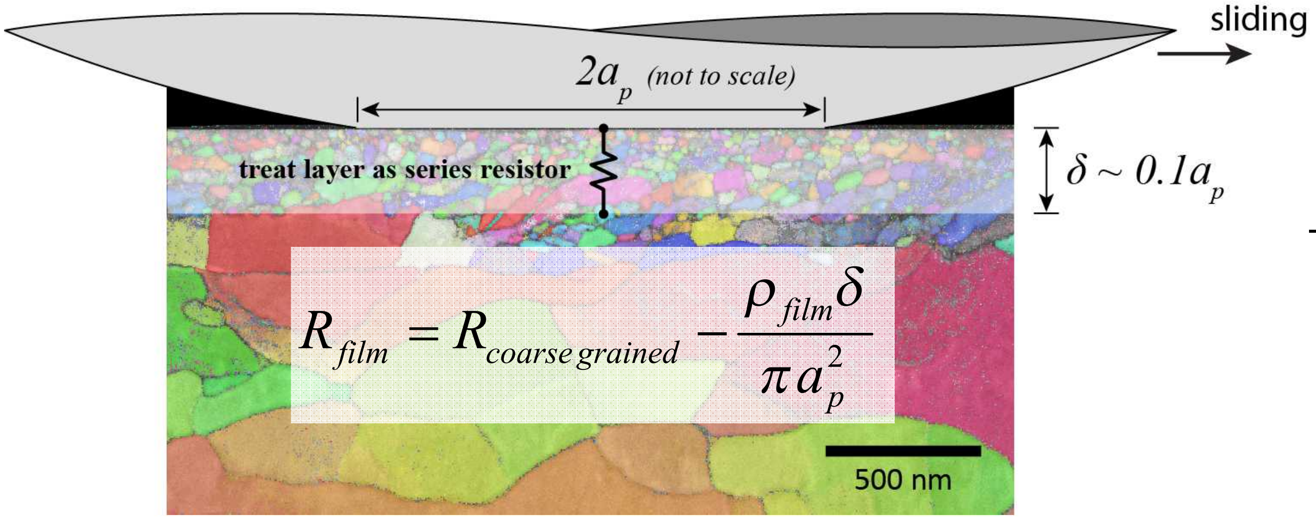
Self-mated pure Cu – we can force low μ by reducing temperature!



$F_n = 100$ mN
1.6 mm radius

Low μ achieved by reducing temperature at conditions that produce high μ at ambient temp.

Electrical resistance data provides an in situ indirect measure of grain size in NC film



$$R_{film} = R_{coarse\,grained} - \frac{\rho_{film}\delta}{\pi a_p^2}$$

$$A_p = \frac{F_n}{H} \longrightarrow a_p = \sqrt{\frac{F_n}{\pi H}}$$

$$H_{Cu} \sim 1\ to\ 3\,GPa$$

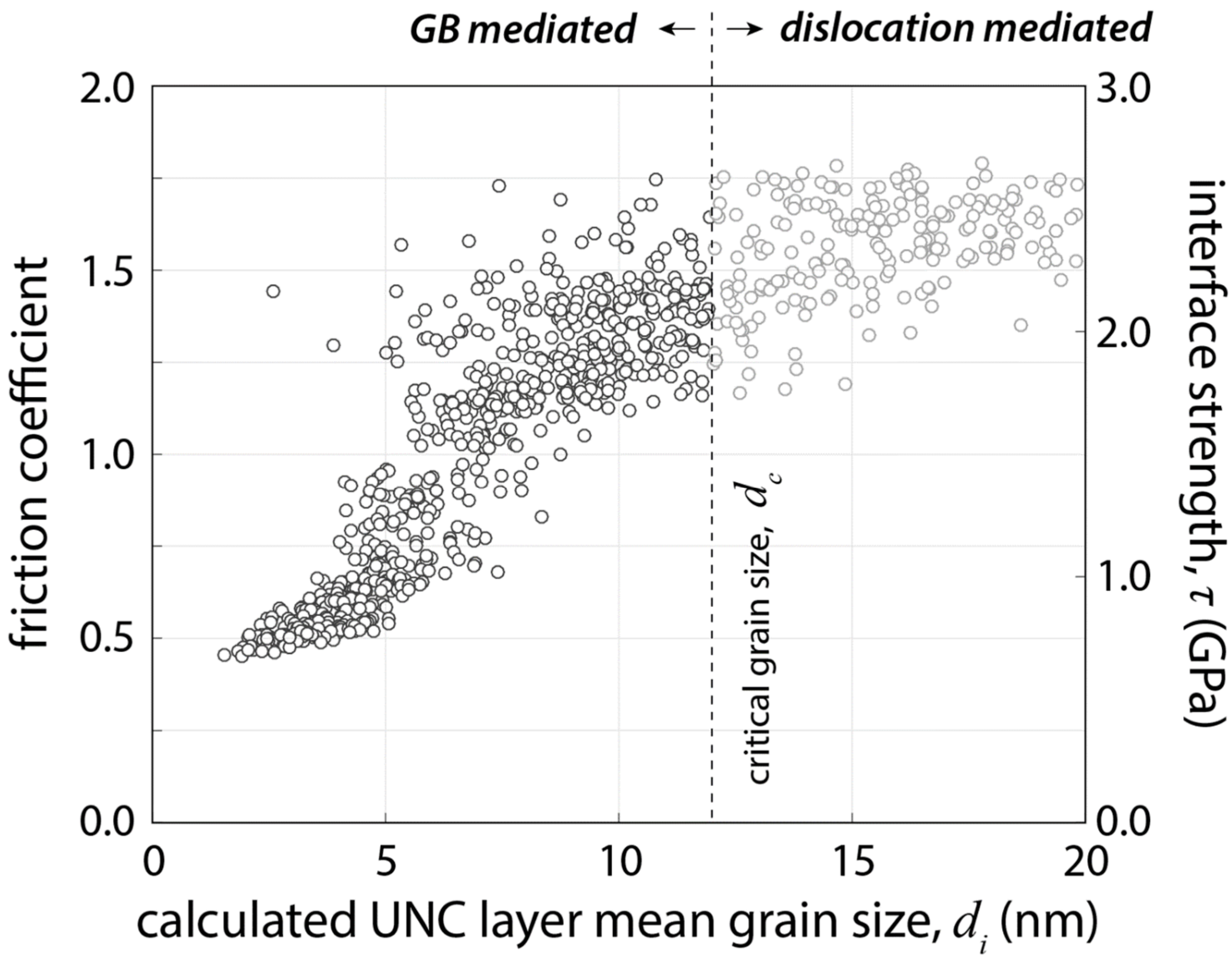
$$a_p \sim 3\,to\,5\,\mu m$$

$$\delta \sim 400\,nm$$

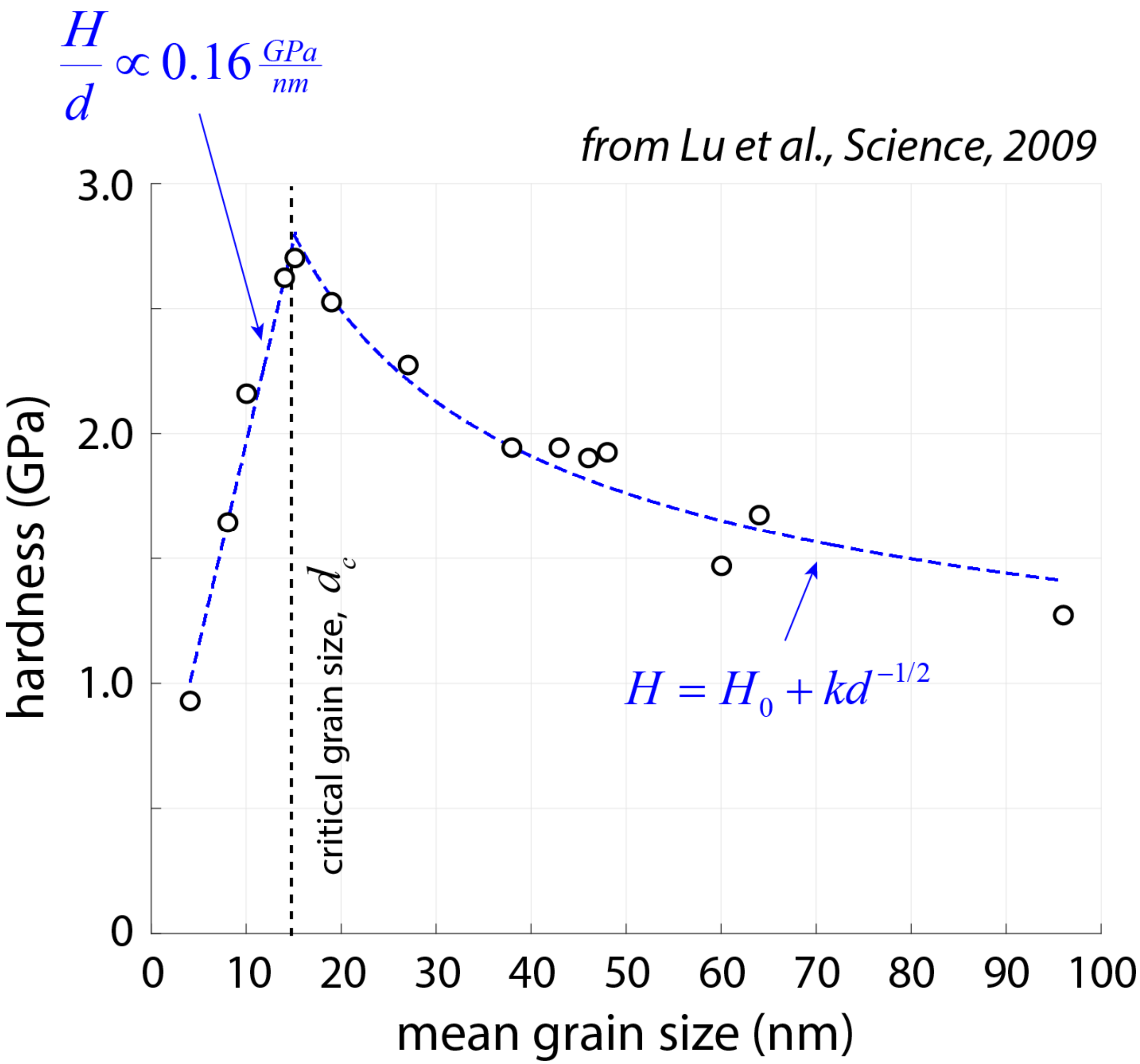
$$d_{film} \cong \frac{4\lambda}{3}\left(\frac{p}{1-p}\right)\left(\frac{\rho_0}{\rho_{film}-\rho_0}\right)$$

Applying Mayadas-Shatzkes electron scattering model described in Argibay, Mogonye, Michael et al., JAP 117, 2015

Now we have a direct connection between friction and grain size!



We have great data from the literature for copper



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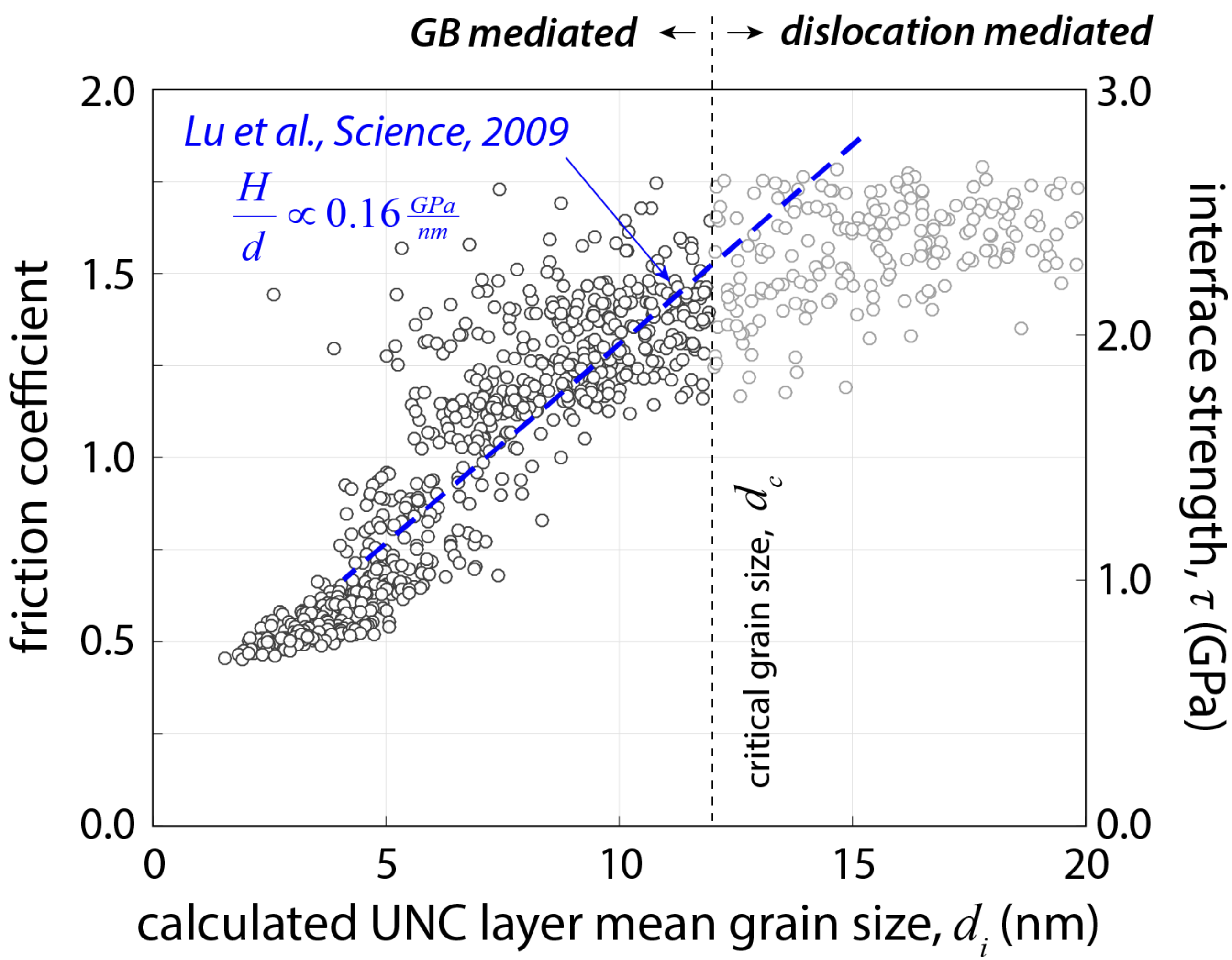
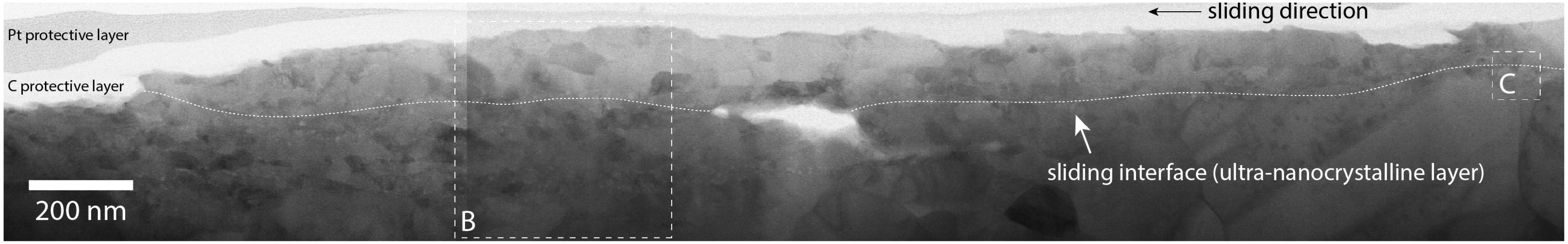
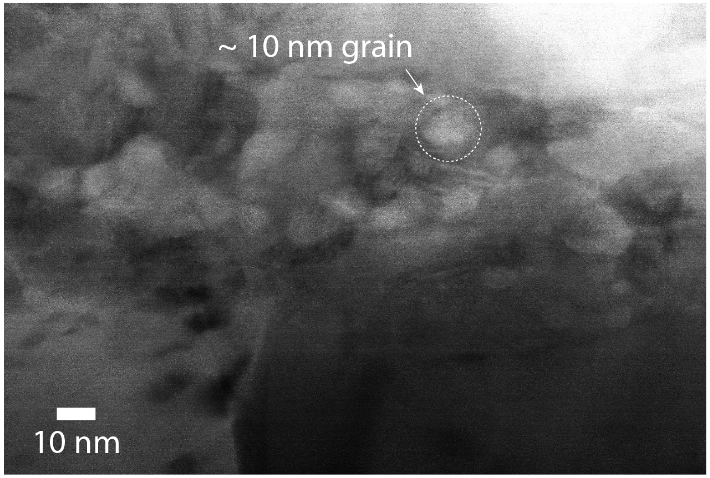


Measured grain sizes for Cu

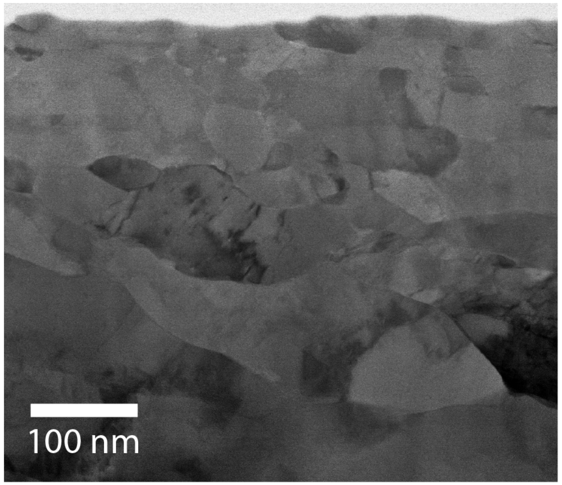


critical grain size: $d_{crit} = 11.8$ nm

$\mu \sim 0.4$



$\mu > 1$



- Grains rapidly coarsen at room T
- Even a remnant of a small grain is a good sign

Conclusions

- Grain size drives friction coefficient for metal-on-metal contacts
  - Low stress/Low temperature = UNC with fine grain size and persistent low friction
  - High stress/High temperature = coarse grained and high friction
- Friction behavior follows inverse Hall-Petch trend below critical grain size
  - *Reduced* interface shear strength of UNC layer
  - Transition from dislocation to grain boundary mediated plasticity

Supplemental Slides

Low friction has been seen in pure metals in the past

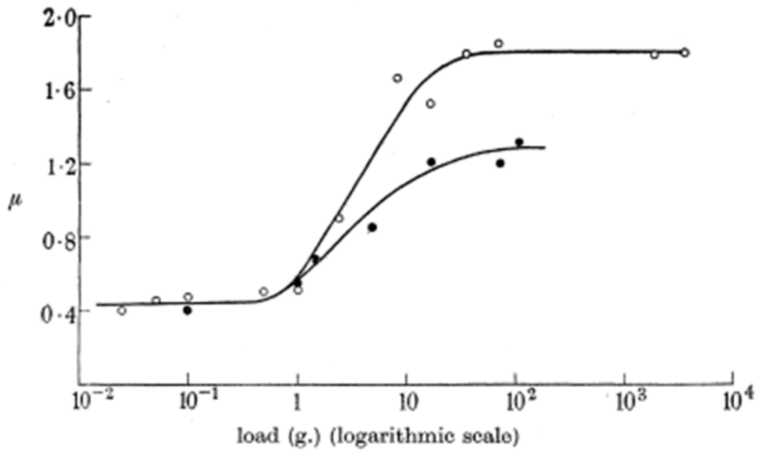
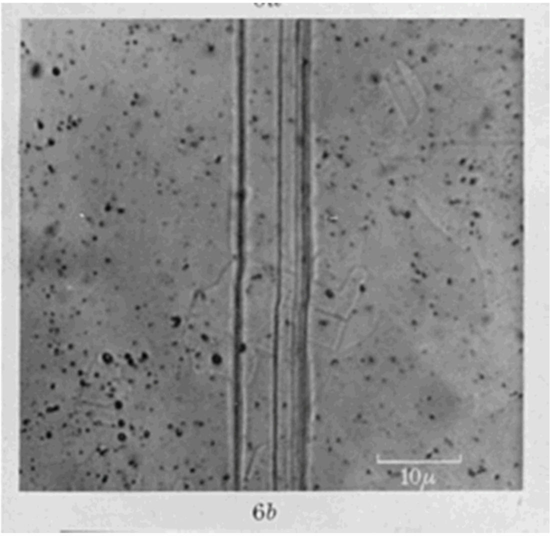


FIGURE 5. The friction of copper on electrolytically polished copper. ○, freshly prepared; ●, oxidized 400 to 600 Å.

Whitehead, *Proc. Royal Soc*, 1950



Cu @ 10 mN, μ ~ 0.4

"On the other hand… when surfaces are cleaned in a good vacuum, the sliding friction…becomes vanishingly small." Bowden & Hughes, *Nature* 1938.

"It was found quite unexpectedly that with some metals, very low friction less than 0.10 was observed… Gold, silver, copper, and platinum exhibit low friction. However, surface-oxidized copper did not." Tamai, *J. Appl. Phys*. 1961

Two routes to stabilize nanocrystalline metals – kinetic and thermodynamic

ref: Simoes et al., Nanotech. (2010)

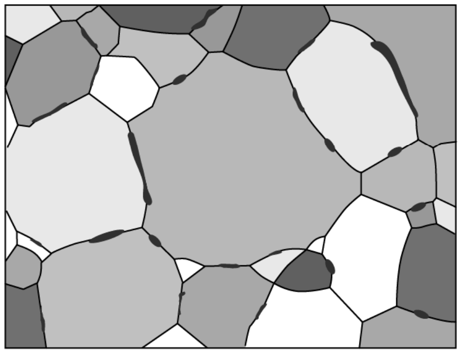
Grain growth is essentialy driven by grain boundary described by speed of grain boundary motion (speed), v →

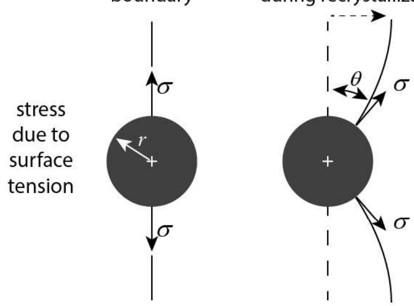
$$v = \boxed{M} \cdot \boxed{P} = \boxed{M_o \exp\left(-\frac{Q_m}{kT}\right)} \cdot \boxed{\frac{2\gamma_o}{r}}$$

M = grain boundary mobility
P = pressure on grain boundary
γ_o = interfacial energy per unit area
r = mean grain radius

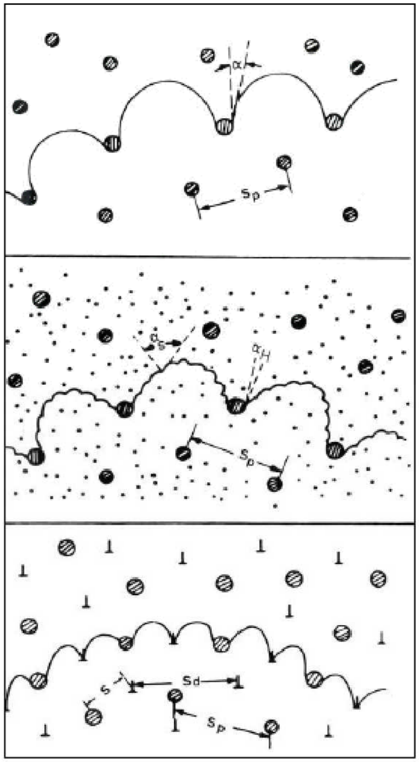
Limit the ***kinetics*** of recrystallization (traditional quasi-stability)

e.g. Zener pinning, solute drag, porosity





drag force: $f_D = 2\pi r \sigma \cos\theta \sin\theta$



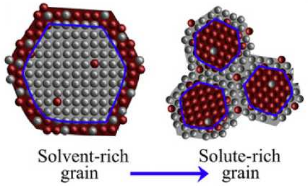
Weissmüller (1993), Kirchheim (2002), and Schuh (2012) have made significant contributions toward understanding and achieving ***thermodynamic*** stability by lowering grain boundary energy through solute segregation

Regular Nanocrystalline Solution (RNS) Model:
ref: Chookajorn et al., Science, 2012

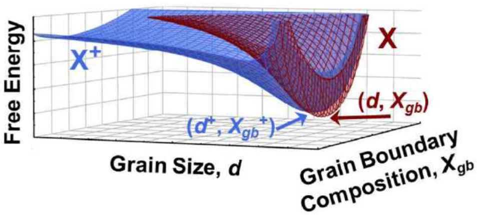
$$\Delta G^{mix} = (1 - f_{gb})\Delta G_c^{mix} + f_{gb}\Delta G_{gb}^{mix} + z v f_{gb}(X_{gb} - X_c)\left[(2X_{gb} - 1)\omega_{gb} - \frac{1}{zt}(\Omega^B\gamma^B - \Omega^A\gamma^A)\right]$$

$$dG = \left[\gamma - \frac{N_\beta}{A}\Delta G_{seg}\right]dA$$

Grain structure model: segregated 2-phase metal system

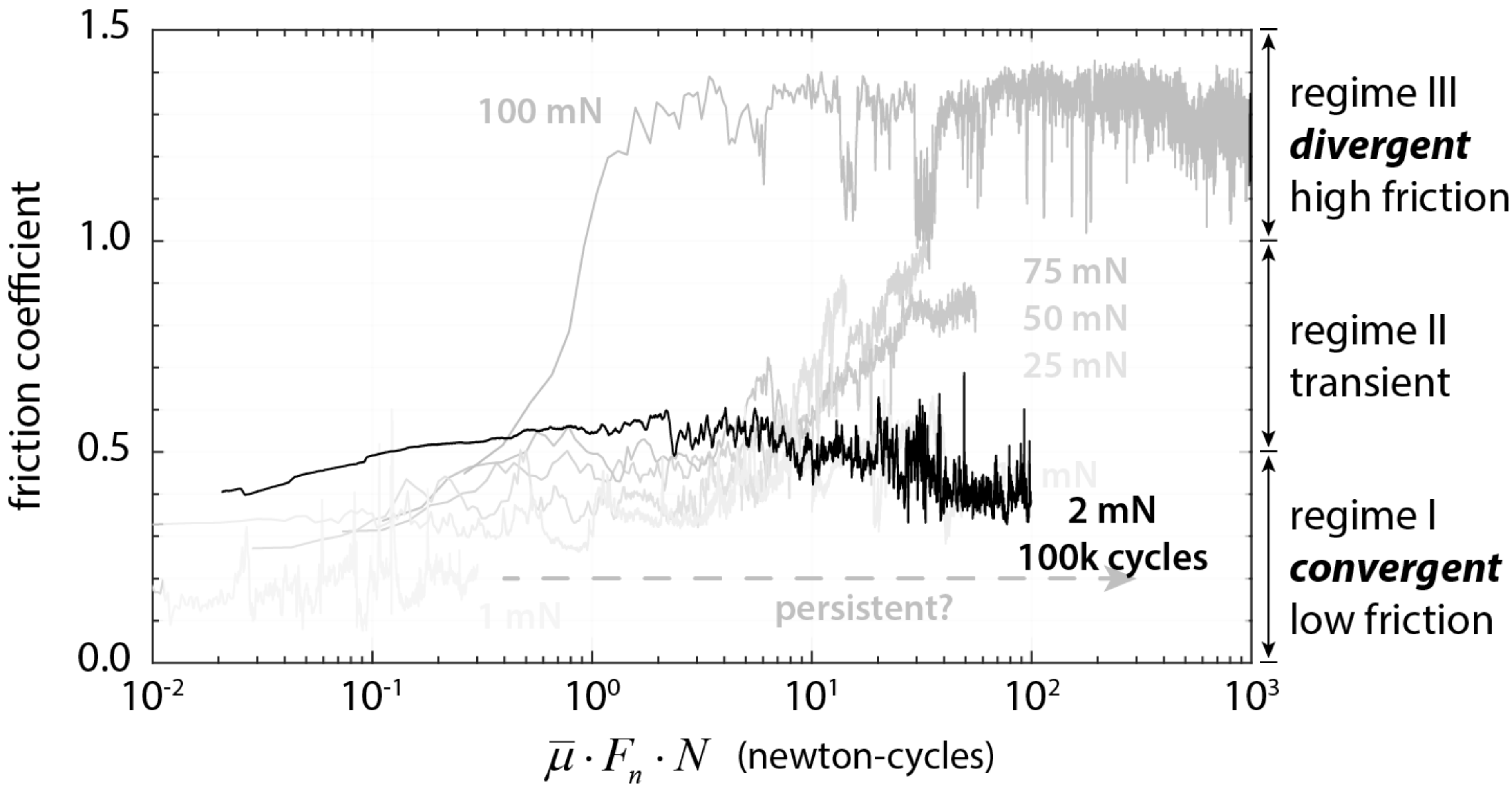


ref: Murdoch et al., Acta Mat. (2013)



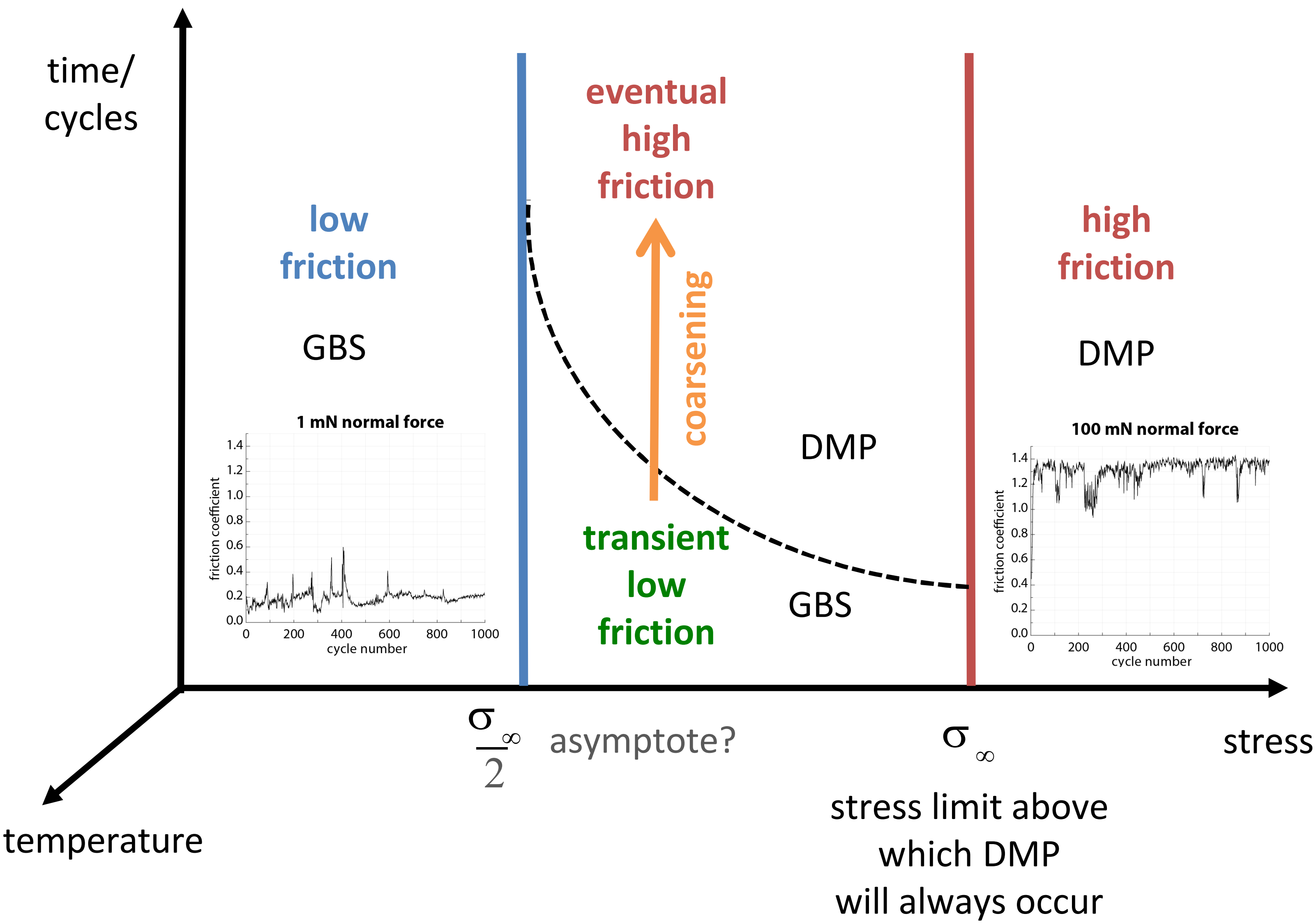
ref: Murdoch et al., Acta Mat. (2013)

So perhaps this is a function of accumulated plastic strain energy… not persistent

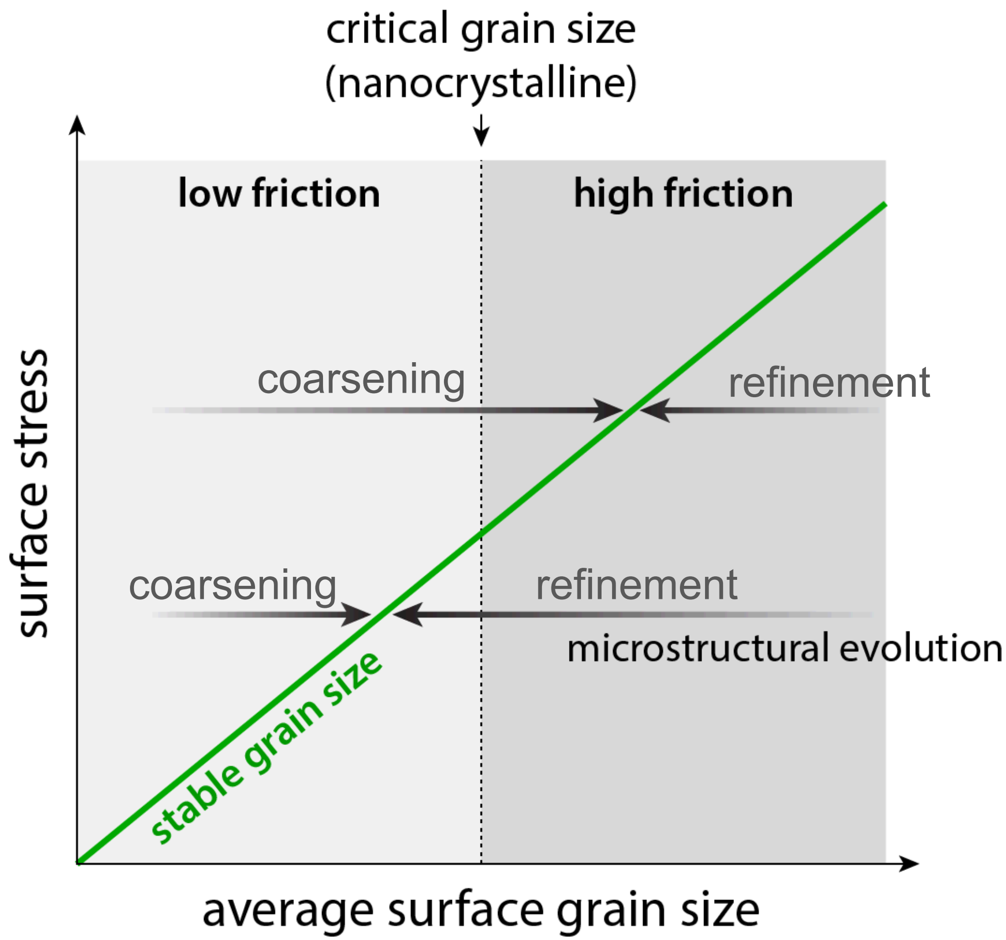


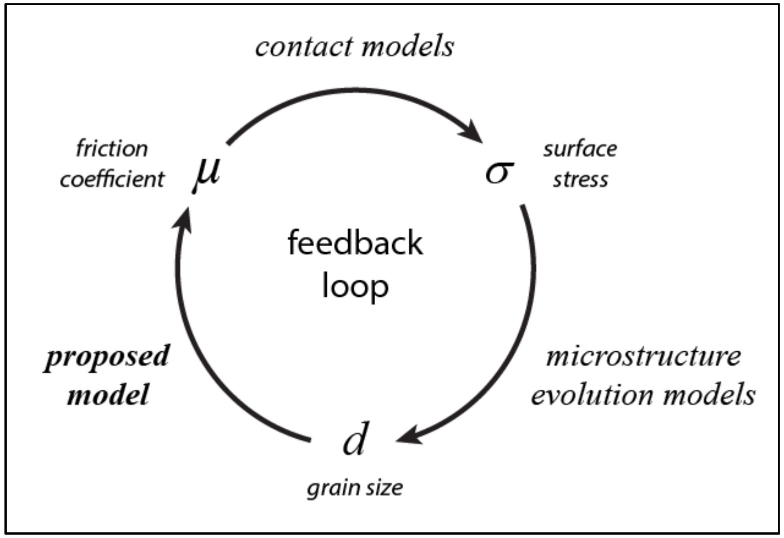
Low friction with pure, soft metals is achievable and PERSISTENT
(i.e. not simply a function of "accumulated damage" or "run-in")

Generalized friction regimes map for metals

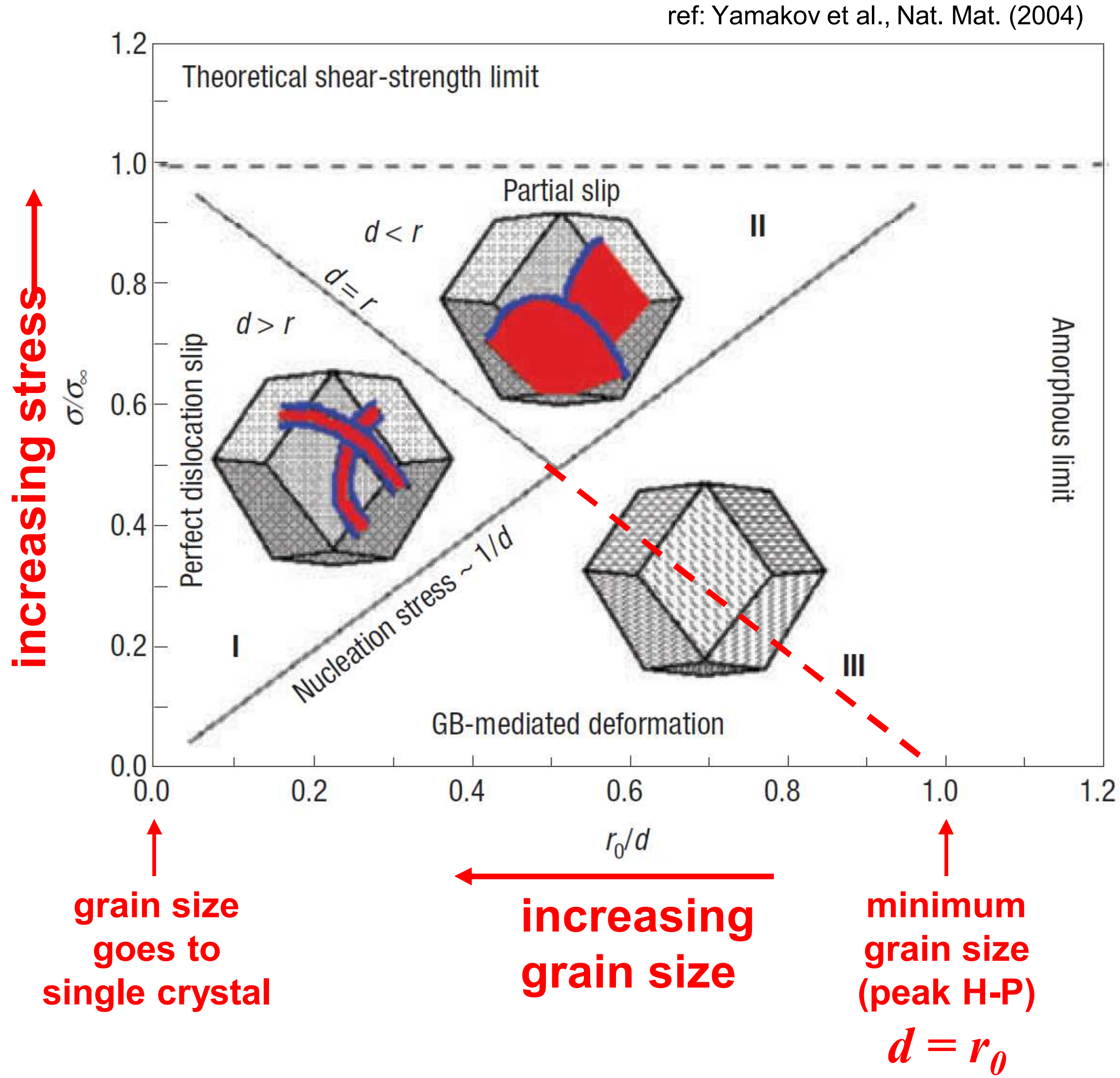


We can predict this critical stress, and therefore the critical grain size





Yamakov et al. (Nat. Mat. 2004) plasticity mechanisms/grain size criteria



Equilibrium (zero stress) dislocation splitting distance:

$$r_0 = \frac{(2+\nu)Gb^2}{4\pi(1-\nu)\gamma_{sf}}$$

Stress-dependent splitting distance:

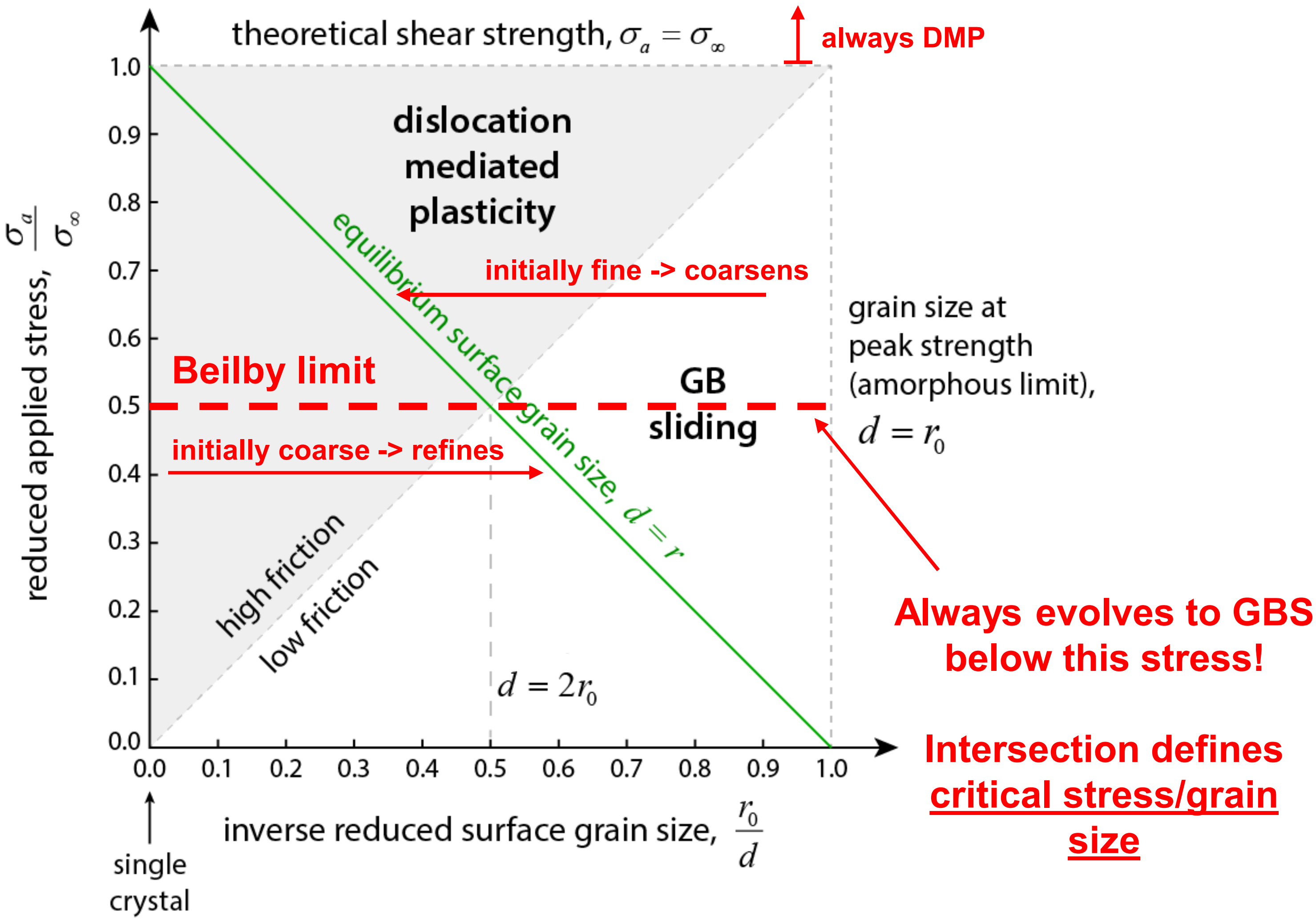
$$r = \frac{r_0}{1-\sigma_a/\sigma_\infty}$$

Theoretical shear strength:

$$\sigma_\infty = \frac{2\gamma_{sf}}{b}$$

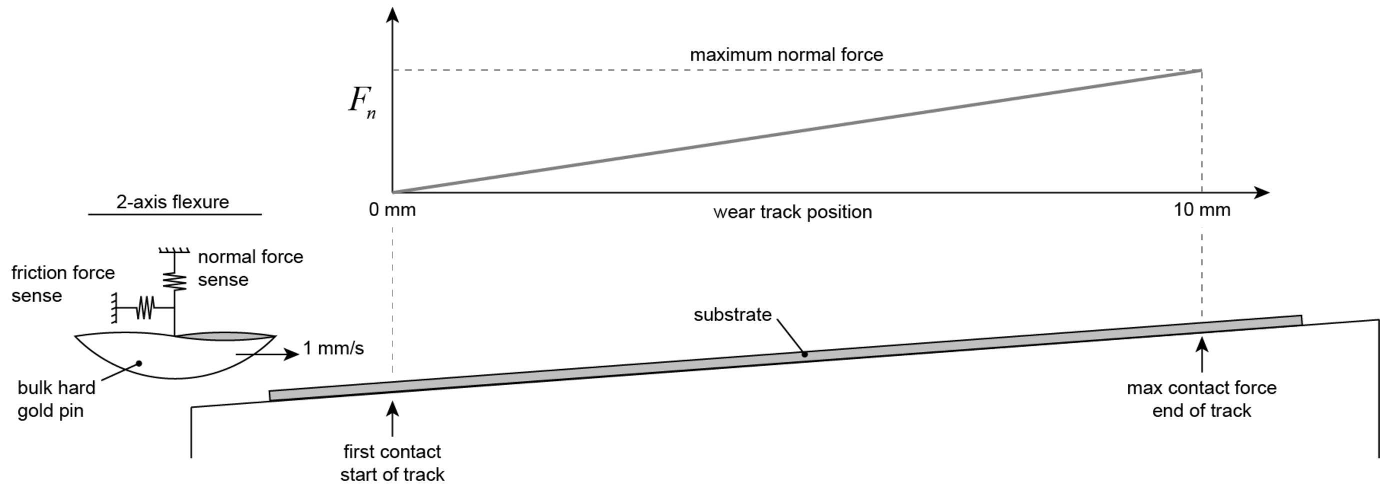
Ref: Froseth et al., Acta Mat. (2004)

Recasting the Yamakov et al. criteria for tribology

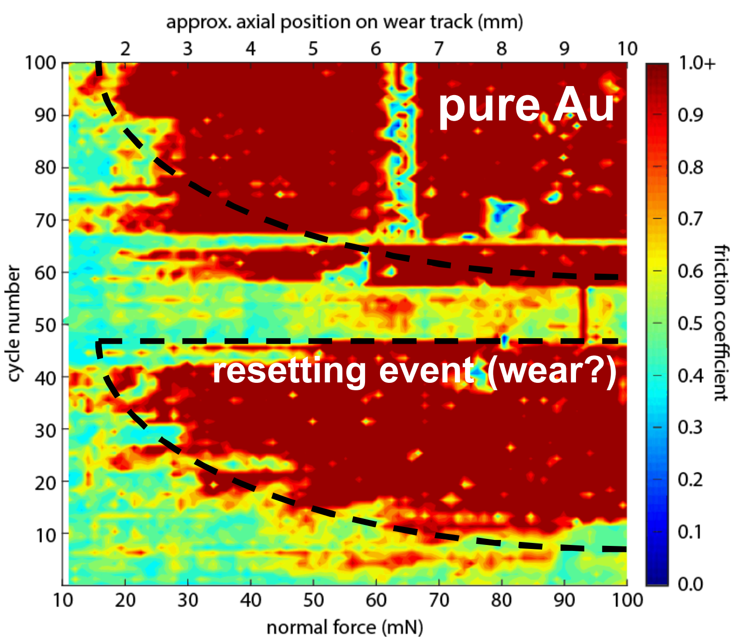


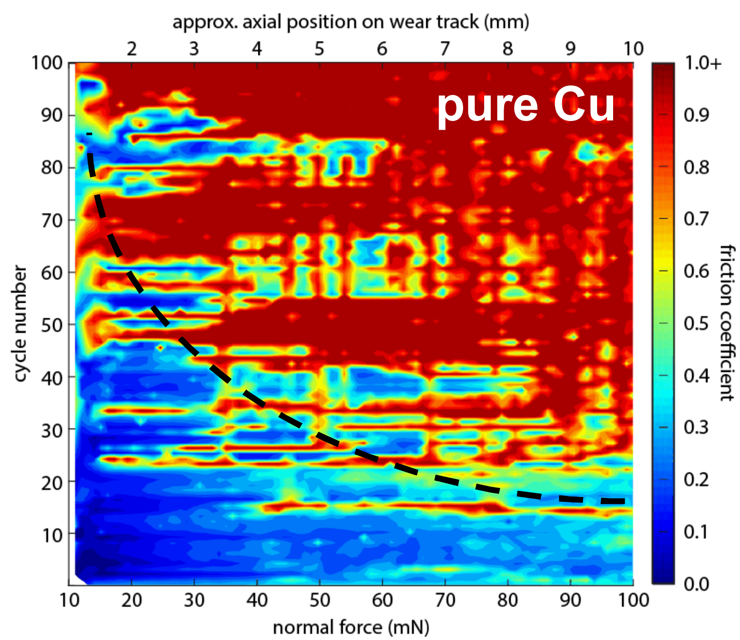
Intersection defines <u>critical stress/grain size</u>

How can we test theory in one shot? Ramped contact force experiments

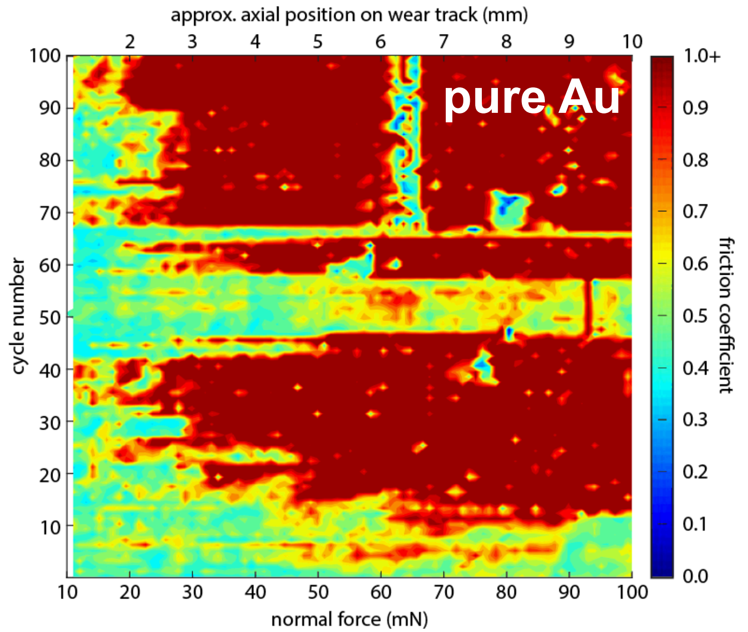


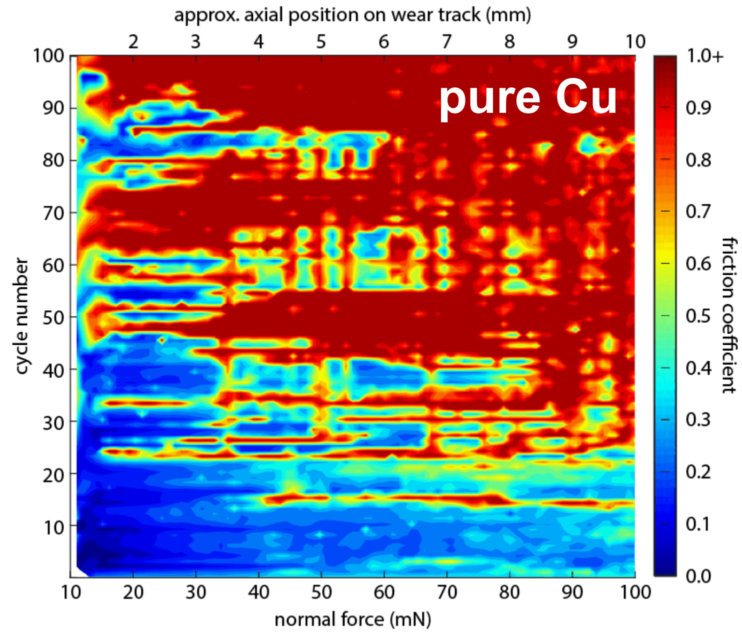
Messy (tribology…), but there is stress-time envelope!

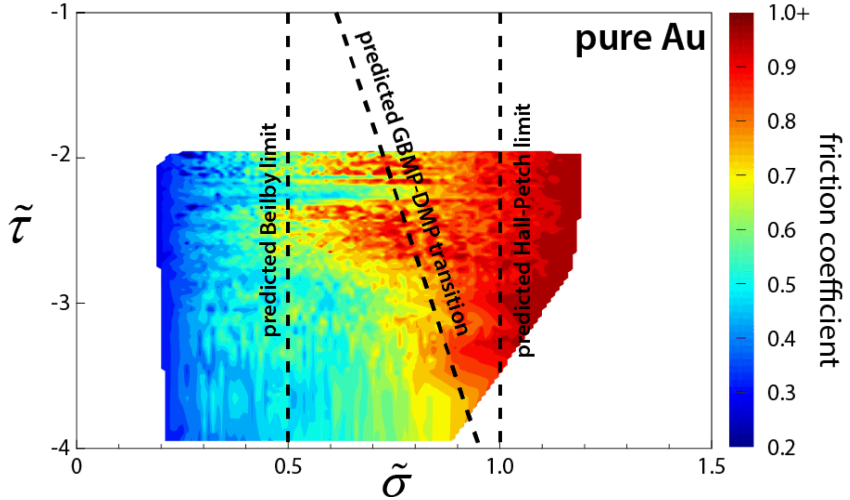


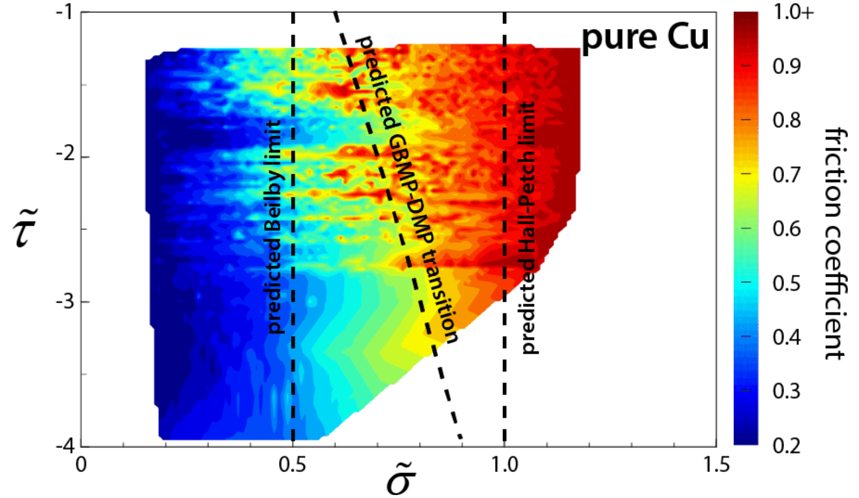


Use Hamilton Stress to Clean Things









Critical grain size predictions *from materials parameters*

$$d_c = 2r_0 = \frac{(2+\nu)Gb^2}{4\pi(1-\nu)\gamma_{sf}}$$

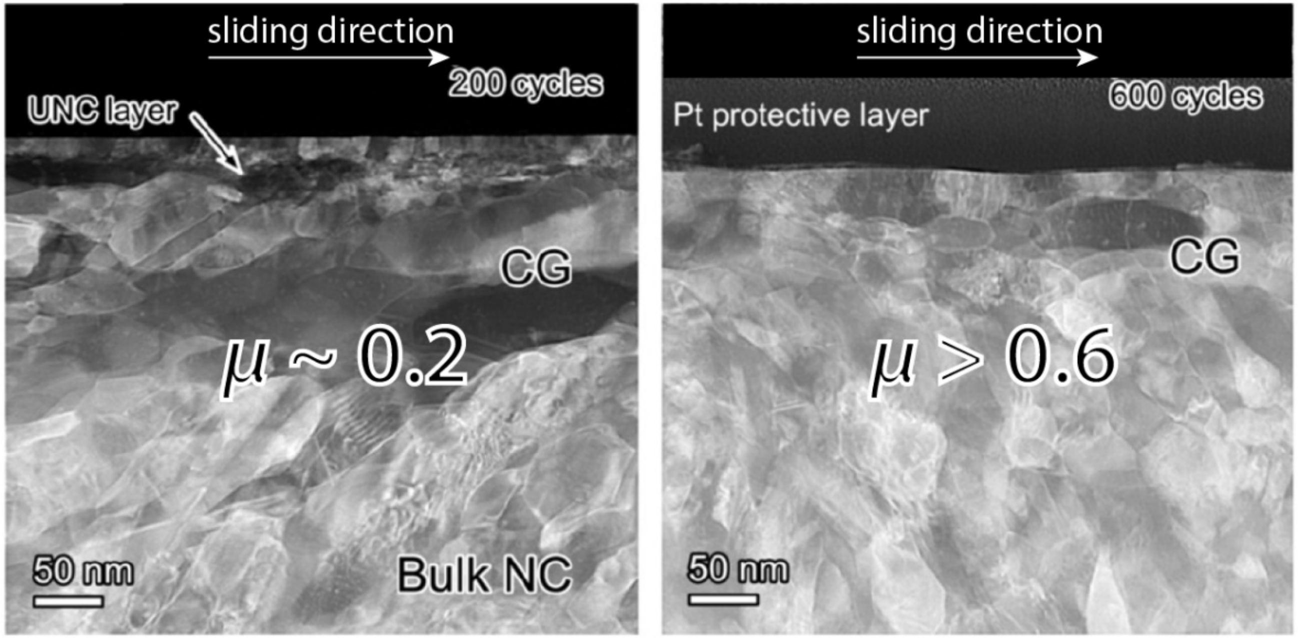
Material	d_c (nm)
Au	17.4
Cu	11.8
Ni	9.8
Al	4.0

This is hard to verify!

- Need low stress at room T
- Stress is localized to the surface
- Very thin layer of very small grains
- Rapid coarsening, even at room T!

Previously reported values for Ni (*Prasad et al., Scripta Mat. 2011*) fit predictions

Low friction UNC layer grain size reported to be 5-10 nm
critical grain size for Ni calculated to be ~ 9.8 nm

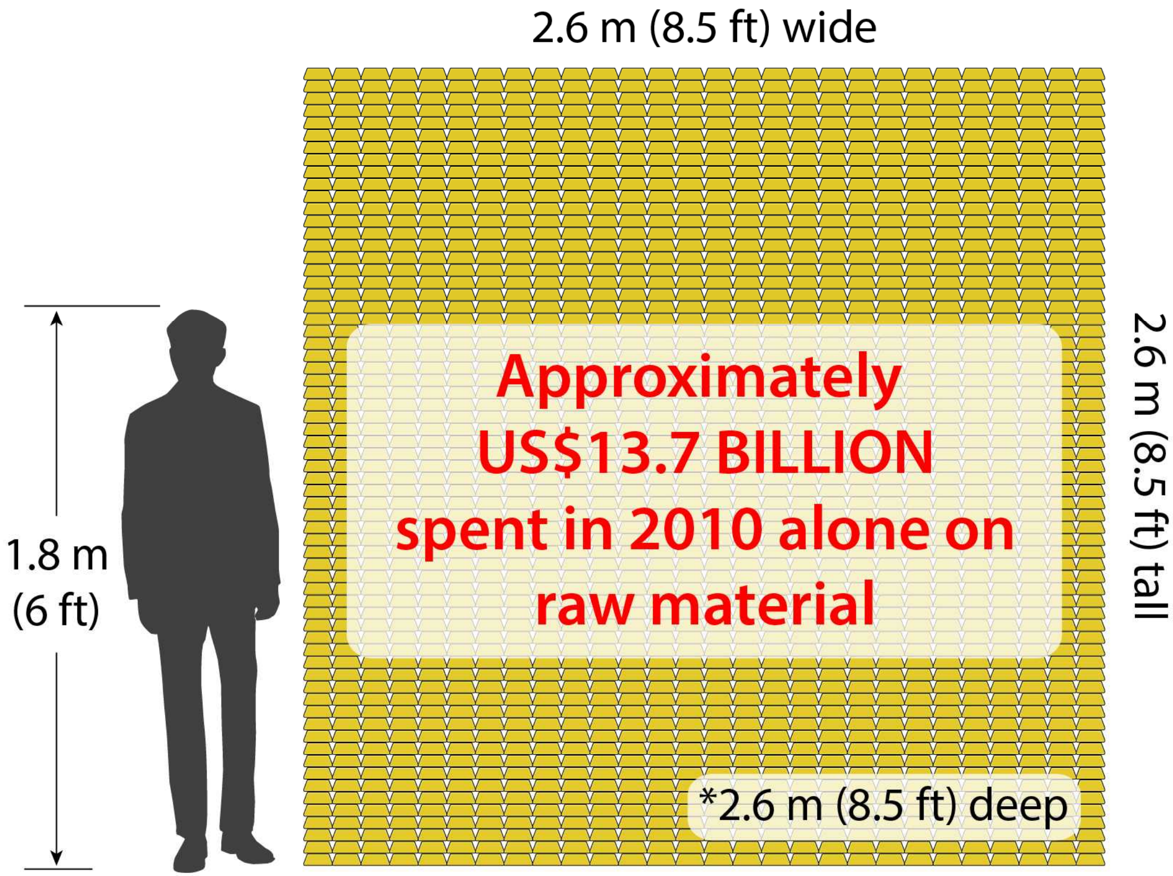


steady-state cross-sections of wear tracks

"The Gold Standard"… how much gold you may ask?   TONS per year

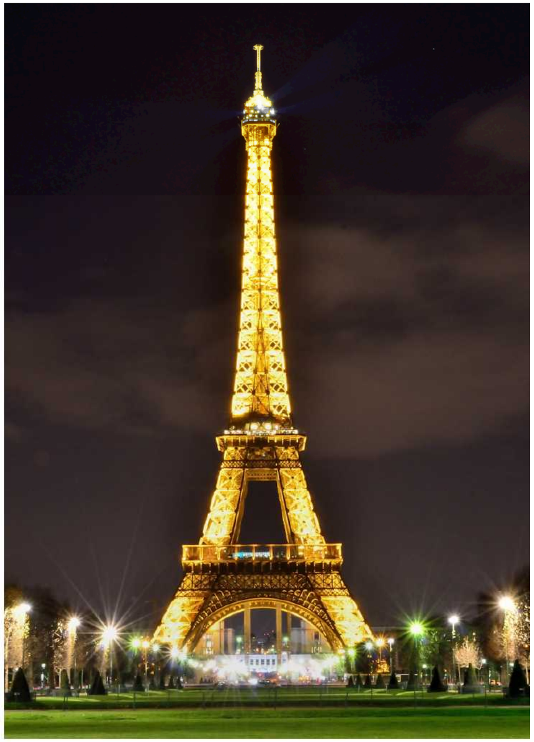
An estimated **300 metric tons/year** of gold used in electronics related applications, most of it in electroplated connectors and contacts (**11% of yearly amount mined**)

Equivalent to a cube comprised of ~25,000 standard gold bars (12 kg/26.4 lb each)…



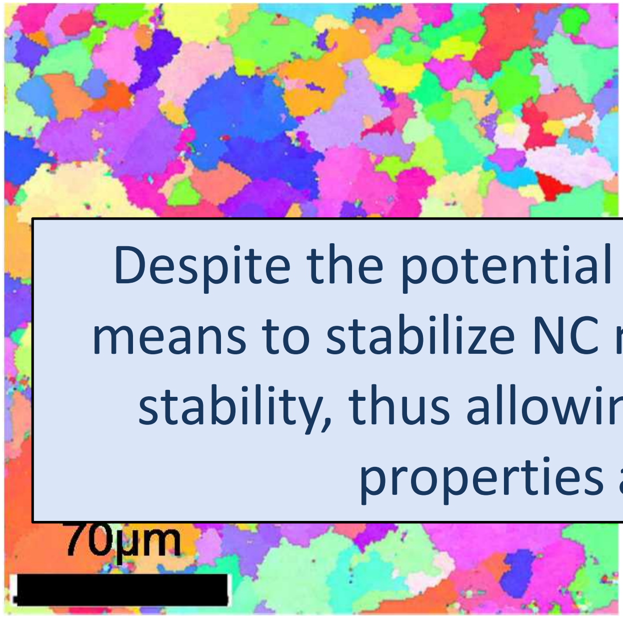
Reference: Gold Survey, Gold Fields Mineral Services Ltd., 2010

… or enough to clad the surface of the Eiffel Tower with 70 µm of pure gold ***every year***



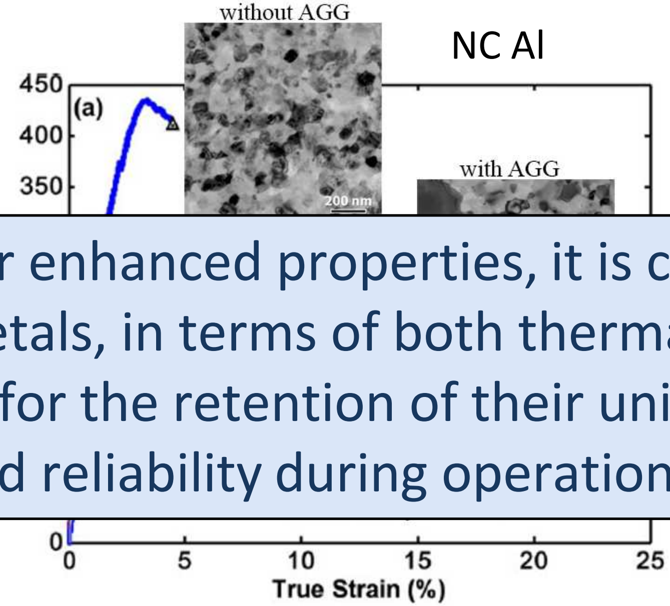
But… NC metals show a propensity for both **thermal** and **mechanical** grain growth at relatively low homologous temperatures (even room temp)!

NC Pd (*originally 10nm grain size*) after two months at RT



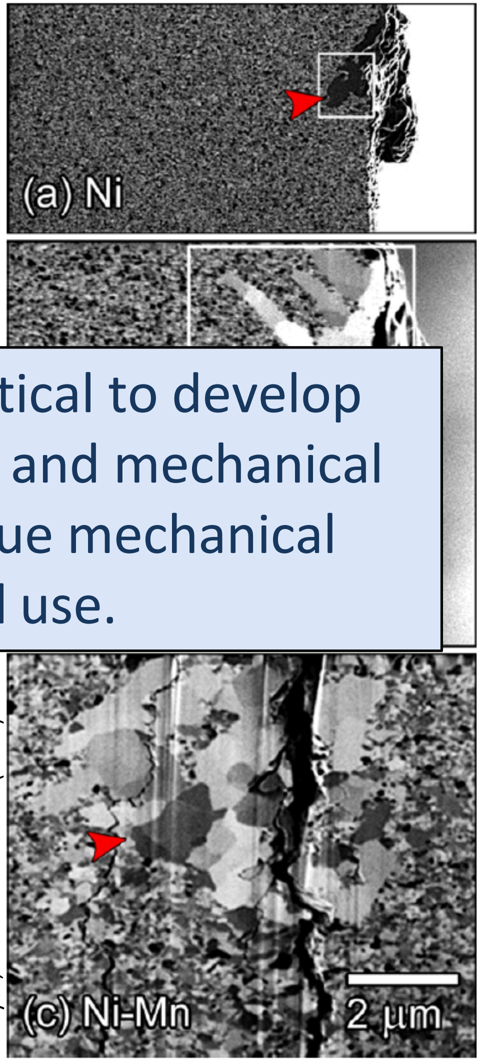
M. Ames, Acta Mater (2008)

Grain growth during **mononotic loading** leading to decreased strength



Gianola et al., Acta Mater. (2006)

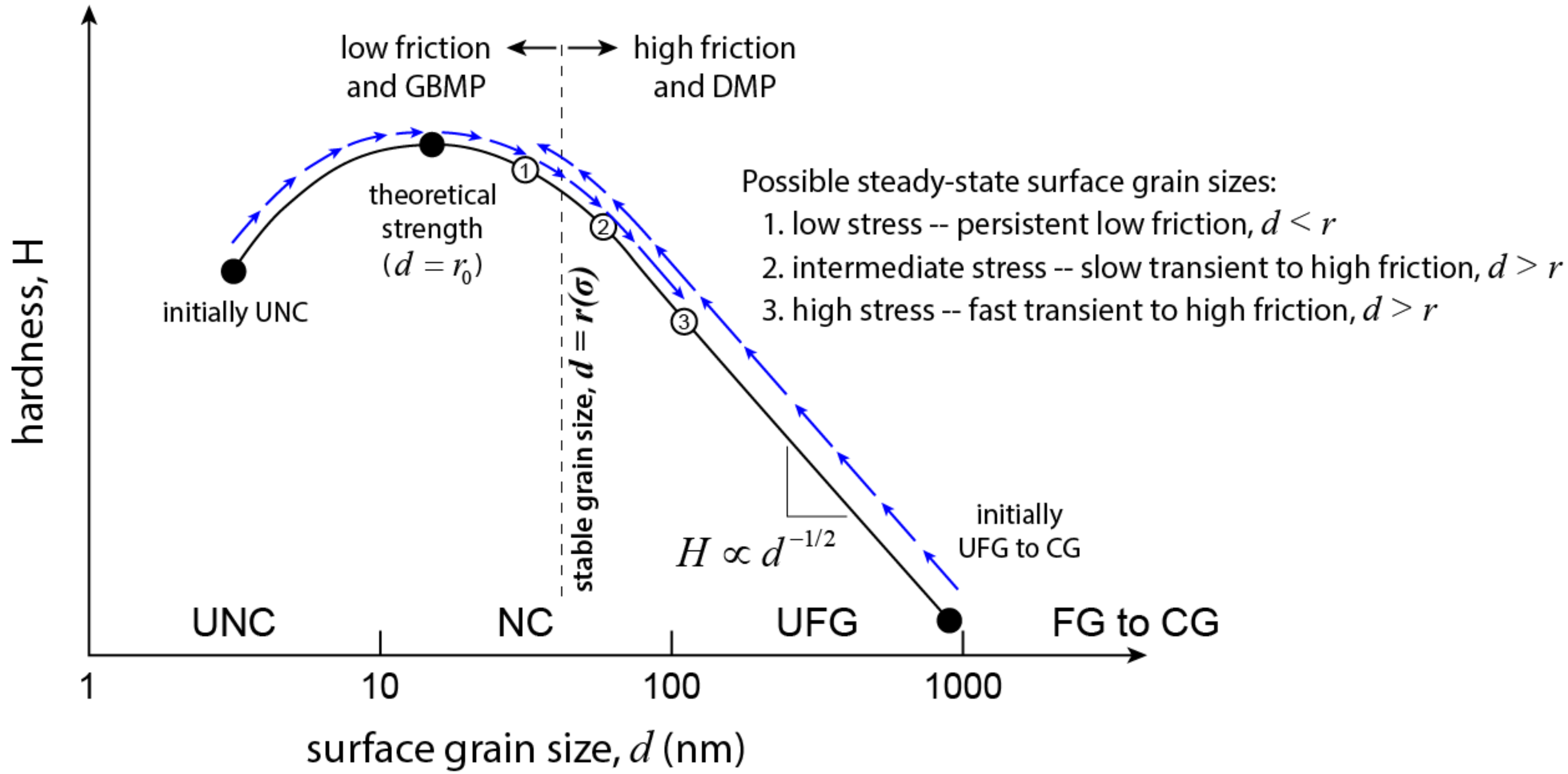
Fatigue-induced grain growth leading to crack initiation



Boyce, Met. Mat. Trans. A (2011)

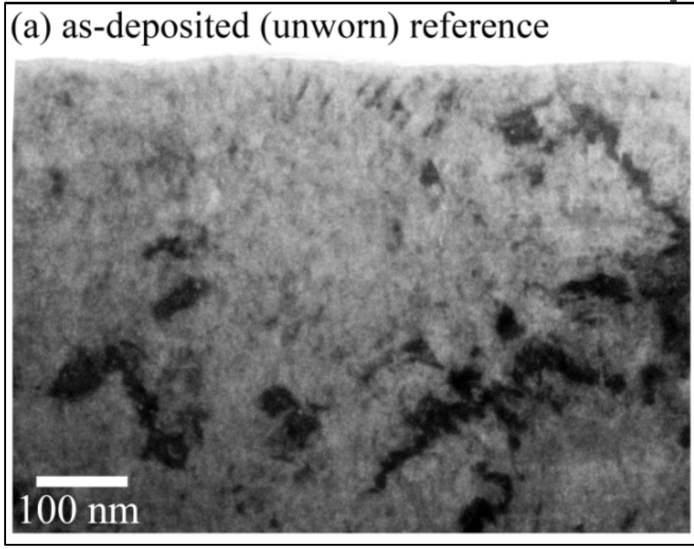
Despite the potential for enhanced properties, it is critical to develop means to stabilize NC metals, in terms of both thermal and mechanical stability, thus allowing for the retention of their unique mechanical properties and reliability during operational use.

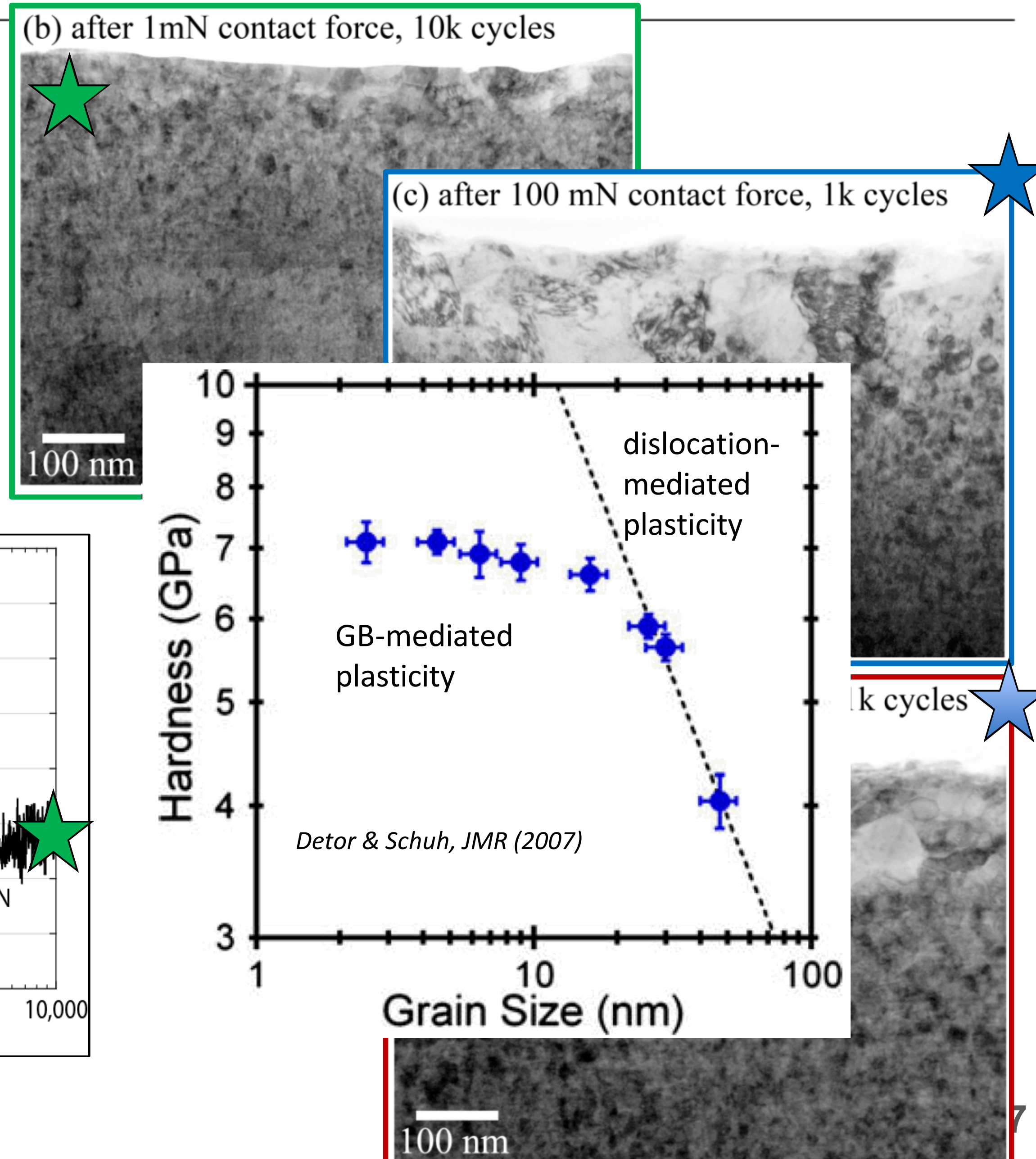
Microstructural Evolution Model explains the Hardness

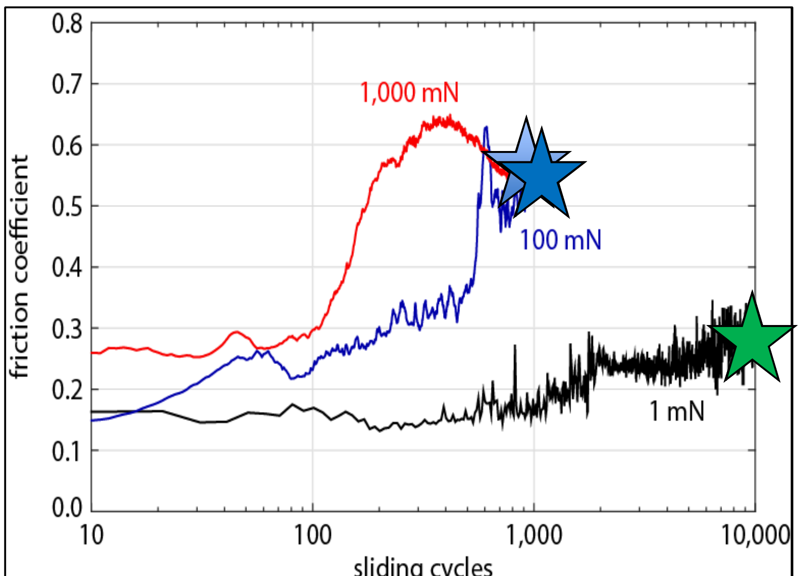


- Initial grains ~ 5 nm
- Model predicts friction transition at ~ 20 nm grains
- Start of in inverse HP regime, coarsen to stable grain size in conventional HP regime

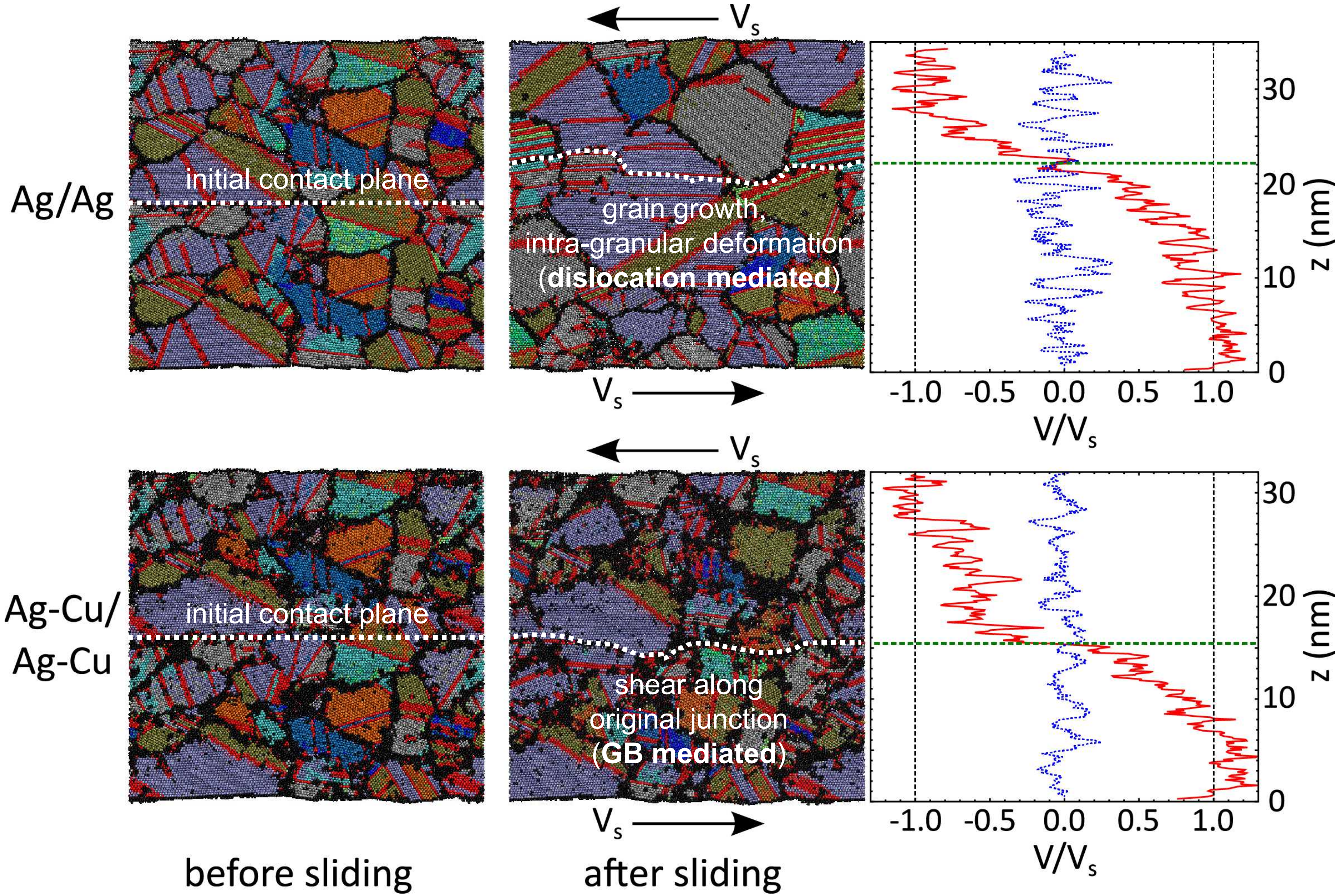
TEM corroborates this interpretation



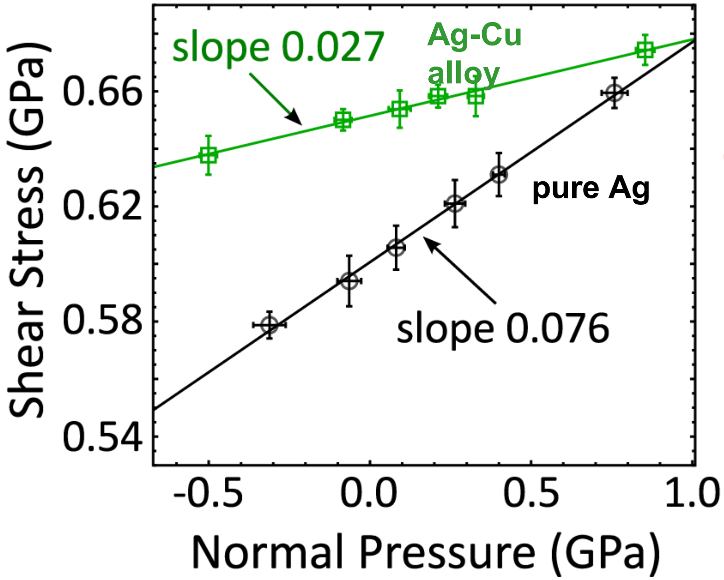




Non-rigid slab-on-slab – two different deformation mechanisms



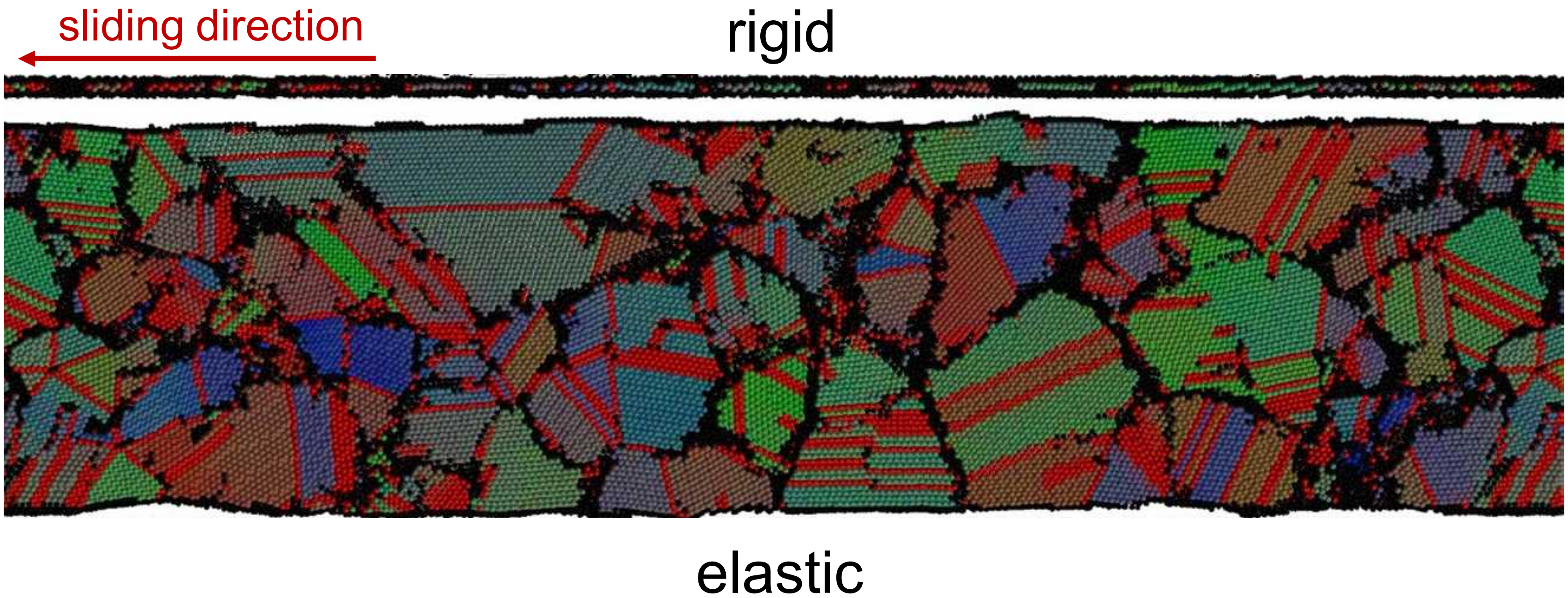
Summarizing findings of MD simulations: grain growth leads to high friction



3x higher friction for pure

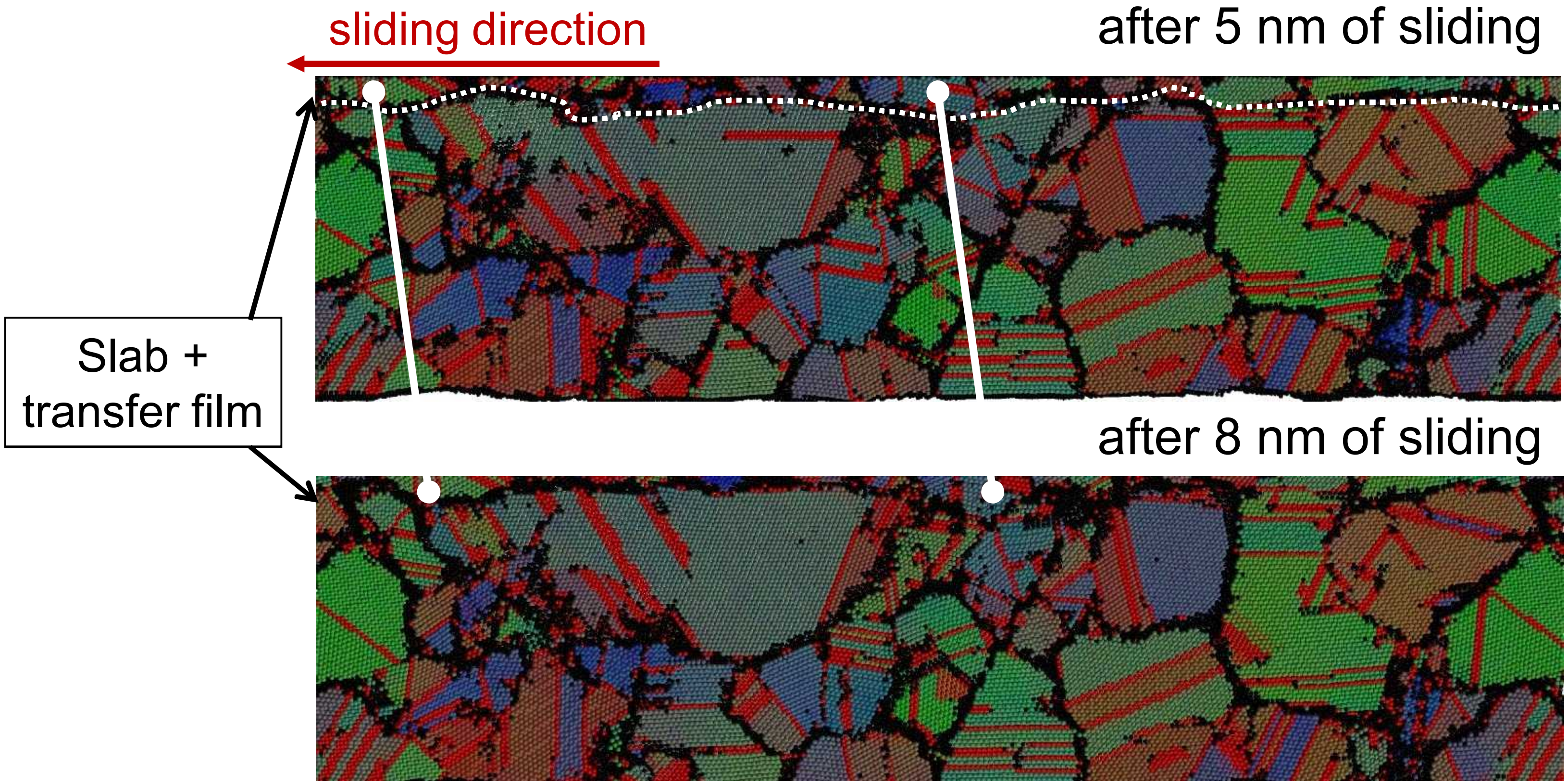
1. Grain growth leads to high friction for:
   a. pure metals
   b. alloys without lattice mismatch
   c. dislocation mediated plasticity
2. Suppressing grain growth leads to low friction:
   a. Alloys with lattice mismatch
   b. Grain boundary sliding (GBS)

We switch to rigid slabs to force grain boundary sliding



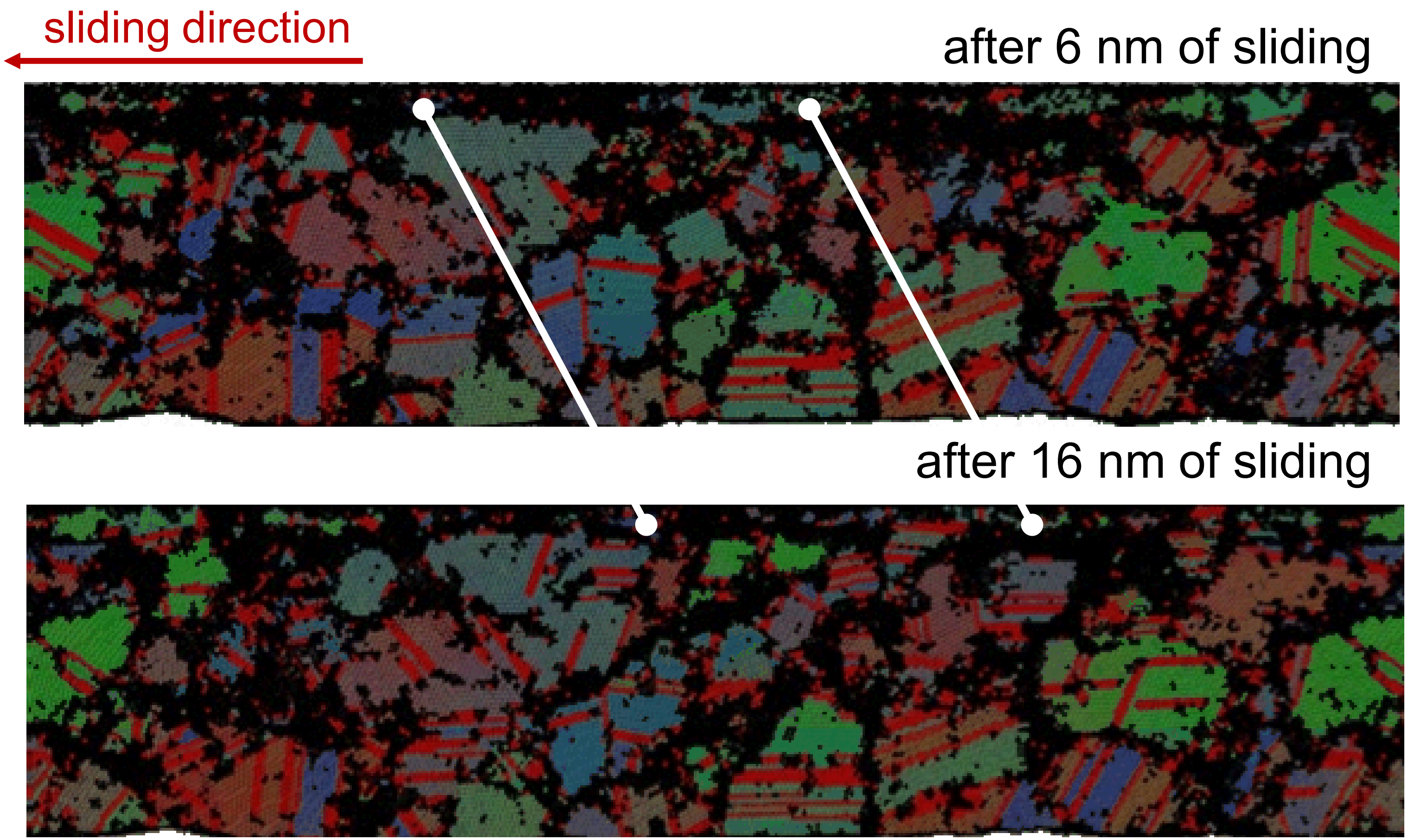
- Rigid slabs suppress grain growth
- No plowing possible, no tip curvature

Sliding of pure Ag slabs



- Forms transfer film
- Grains grow
- Slides along transfer film grain boundaries

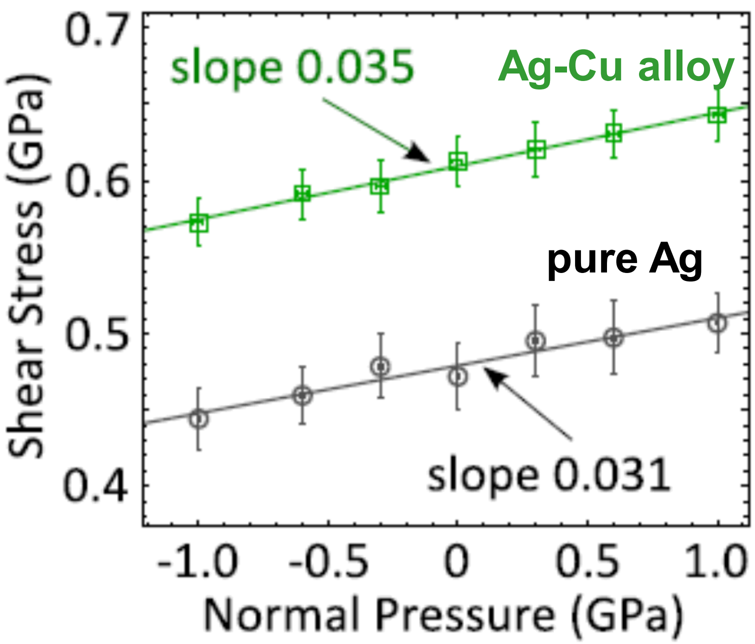
Sliding of insoluble Ag alloy (Ag-10% Cu) -- Cu segregates to/pins grain boundaries



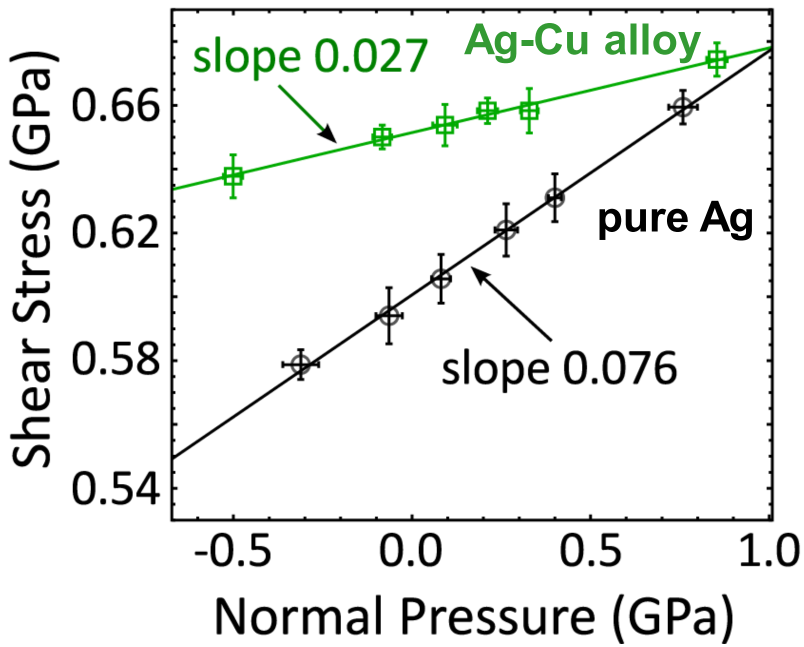
- Alloy ALSO slides at transfer film boundary
- Pure and alloy slabs both shear primarily by GB sliding

μ **essentially identical** – grain growth suppression leads to ***same*** friction mechanism

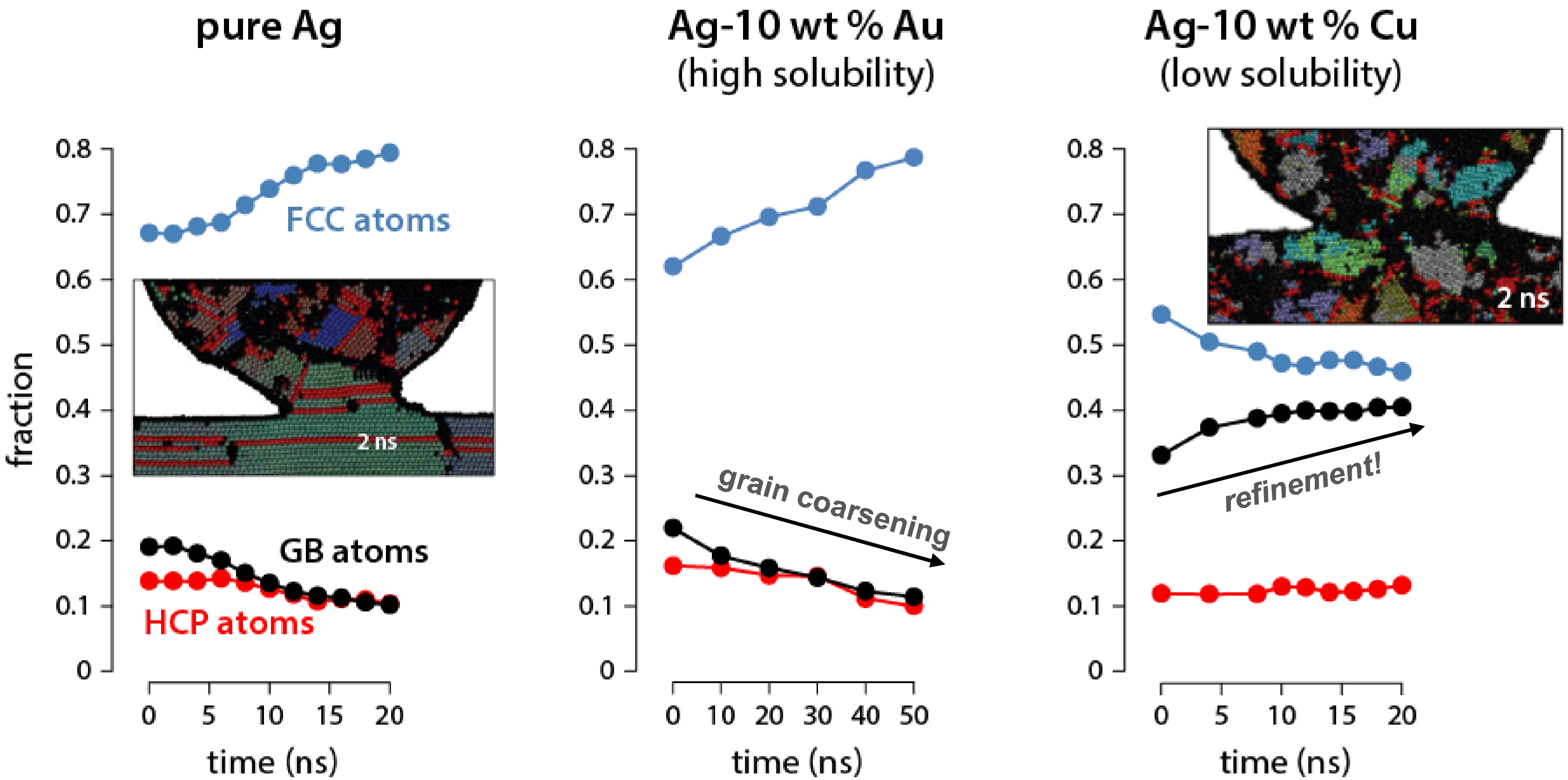
GB Sliding alloy and pure



GB sliding alloy/DMP pure



… alloying (stabilized GBs) changed **dominant deformation mechanism**



high friction & DMP

high friction & DMP

low friction & GBS

What are the predicted grain sizes?

property	material system				
element	Au	Cu	Al	Ni	units
shear modulus, G	27	48	27	76	GPa
Poisson ratio, v	0.44	0.36	0.35	0.31	-
lattice constant, a	4.08	3.61	4.05	3.52	Å
Burgers vector, b	2.88	2.55	2.86	2.49	Å
SFE, γ_{sf}	45	78	166	128	mJ/m^2
GBE, γ_{gb}	378	625	324	-	mJ/m^2
HAGB mobility, M_0	3.84×10^{-6}	30	2×10^{-2}	-	m/s-Pa
activation energy, Q	1.33	2.01	1.05	-	$\times 10^{-19}$ J
calculated parameters					
eq. splitting distance, r_0	8.7	5.9	2.0	4.9	nm
theor. shear strength, σ_∞	312	611	1,117	1,029	MPa
critical grain size, $2r_0$	17.4	11.8	4.0	9.8	nm
critical stress, $\sigma(2r_0)$	156	306	559	514	MPa

Evidence that friction regimes correlate to wear regimes