SAND2020-0704C

New Insights into Alloy Design for Tribological Applications

PRESENTED BY

Dr. Nicolas Argibay

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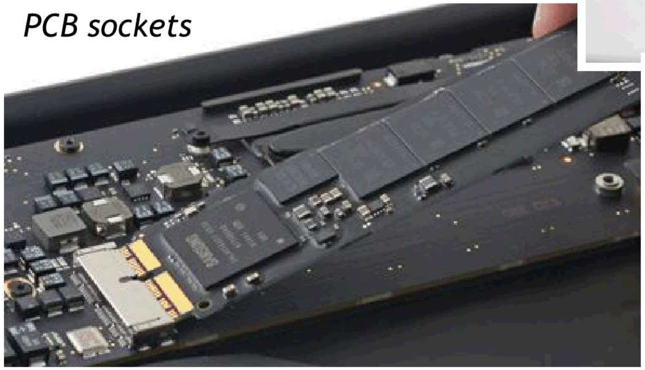
Motivation for Metals Tribology Research

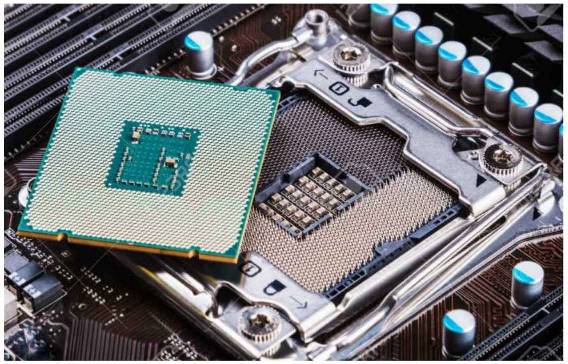
cell phones

wind turbine slip-rings (sensors and blade pitch motors)

PCB sockets



CPU sockets



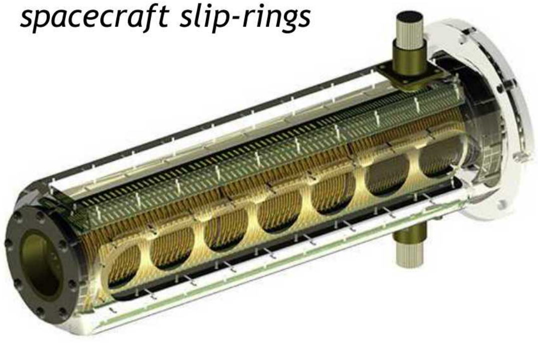
RJ45 connectors

Estimated 150 Metric Tons ($6.9B) of Au used in Electrical Contacts per Year:

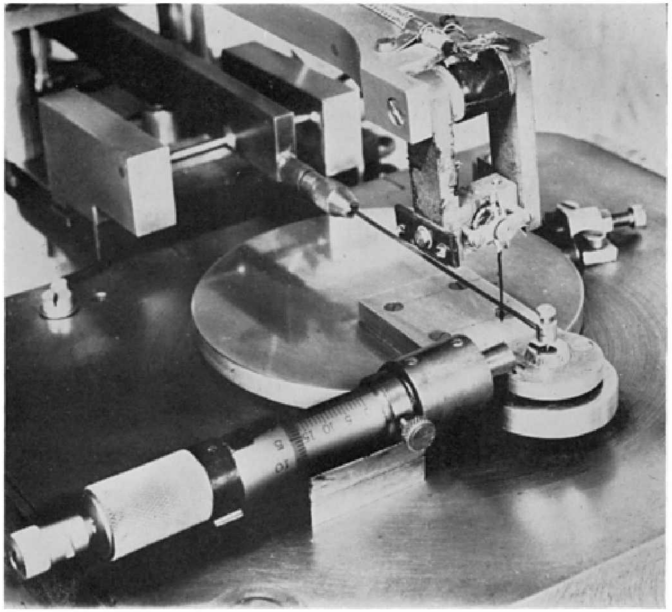
Refs: Gold Survey, Gold Fields Mineral Survey Ltd, 2011
Gold Bulletin 2010, Vol. 43-3, C. Hagelüken and C.W. Corti,
Gold Bulletin 1986, Vol. 19-3, T.D. Cooke

EV charging

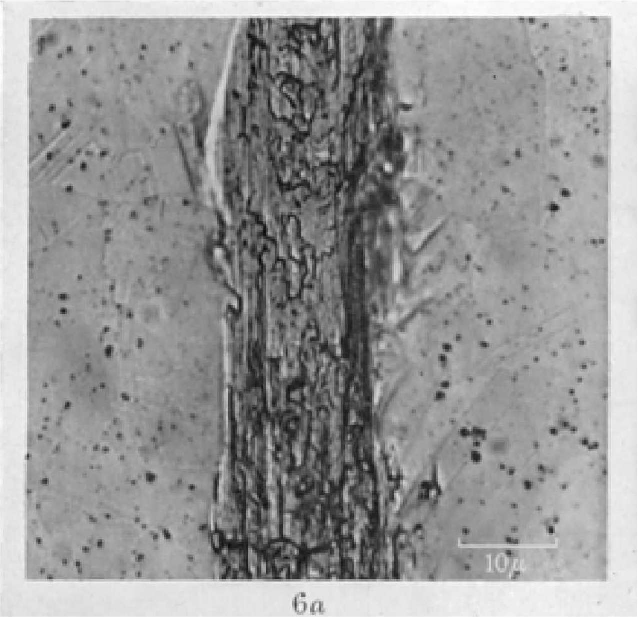
spacecraft slip-rings



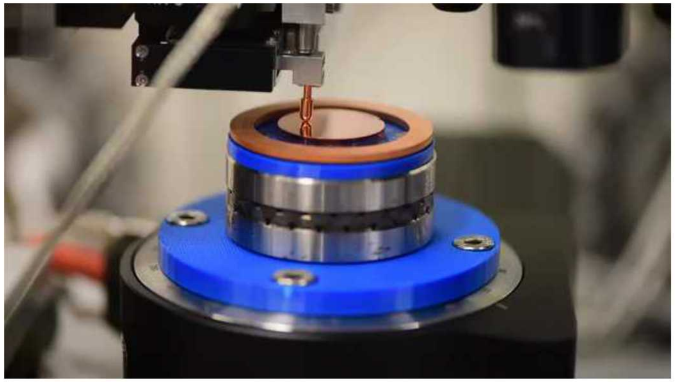
Metals Often Have Poor Tribological Properties

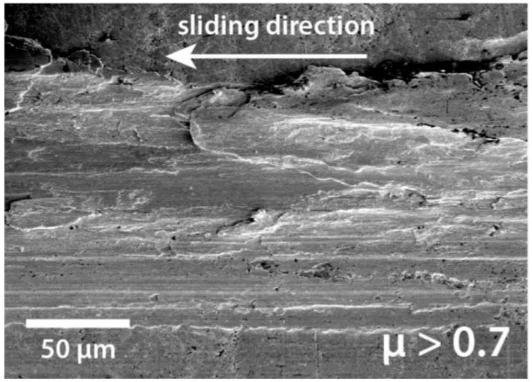


Whitehead, Proc. Royal Soc, 1950



Cu in air @ 150 mN, μ > 1.6

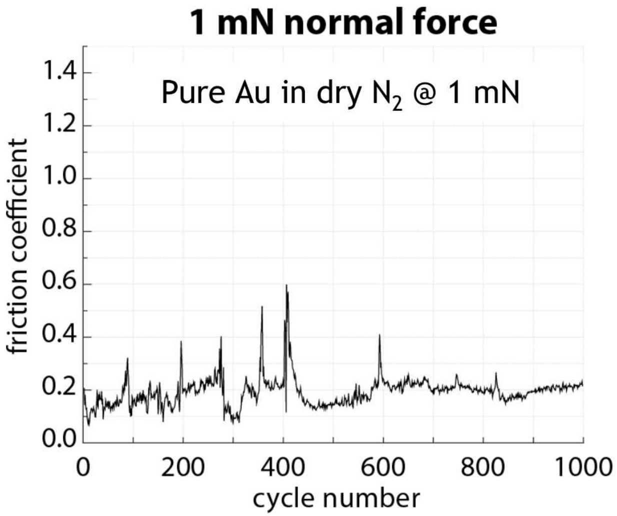




Au in dry N_2 @ 75 mN, μ > 0.7

Metals tend to show strong, rapid interfacial bonding (cold welding) with galling.

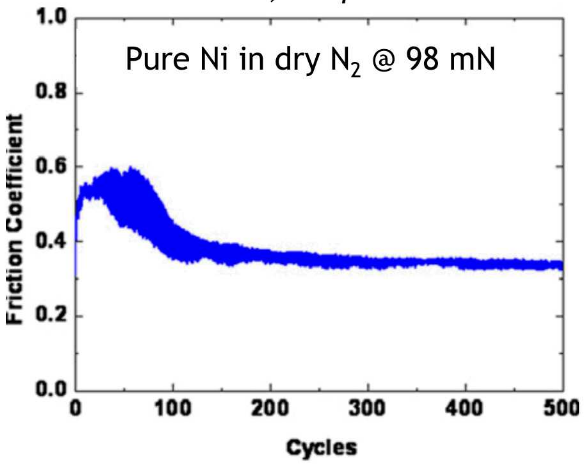
More recent results: Low Friction starting from Bare, Soft, Pure Metals

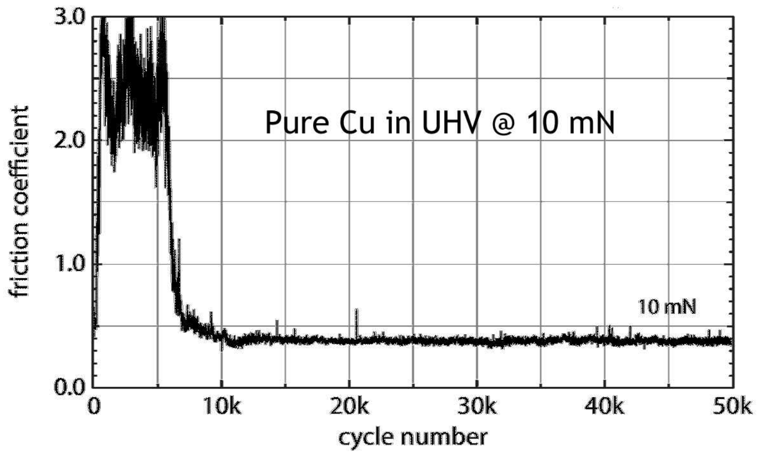


"When surfaces are cleaned in a good vacuum, the sliding friction… becomes vanishingly small."
- Bowden & Hughes, *Nature* 1938.

"It was found quite unexpectedly that with some metals, very low friction less than 0.10 was observed."
- Tamai, *J. Appl. Phys.* 1961

Prasad, *Scripta Mat*. 2011

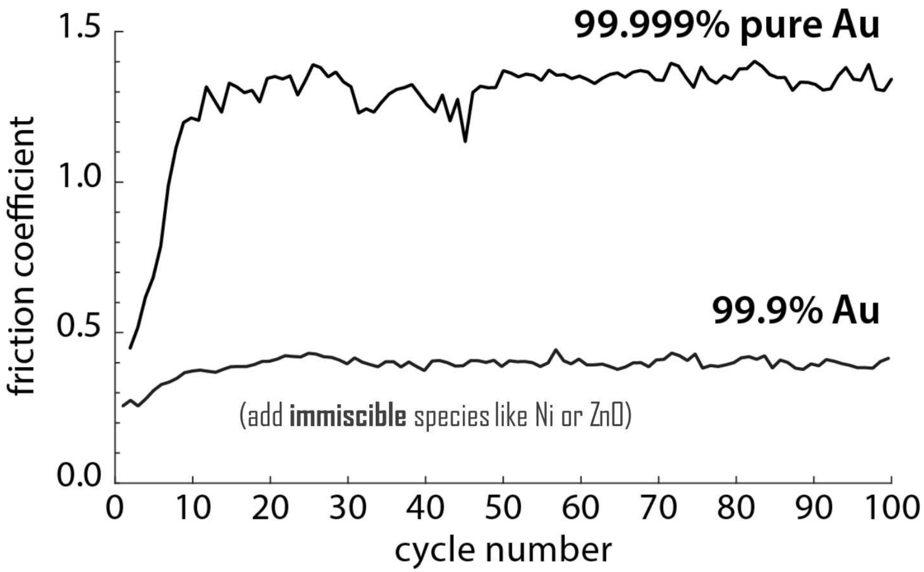




Low friction with pure metals is achievable.

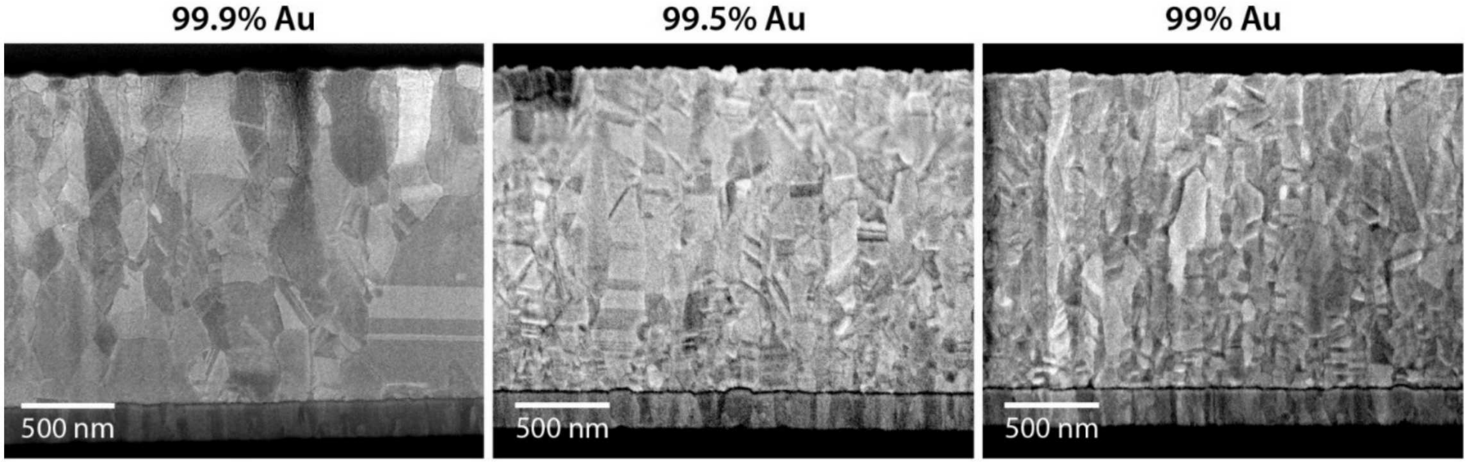
Low Friction and Wear of Nanocrystalline Metals

Alloying reduces friction coefficient:

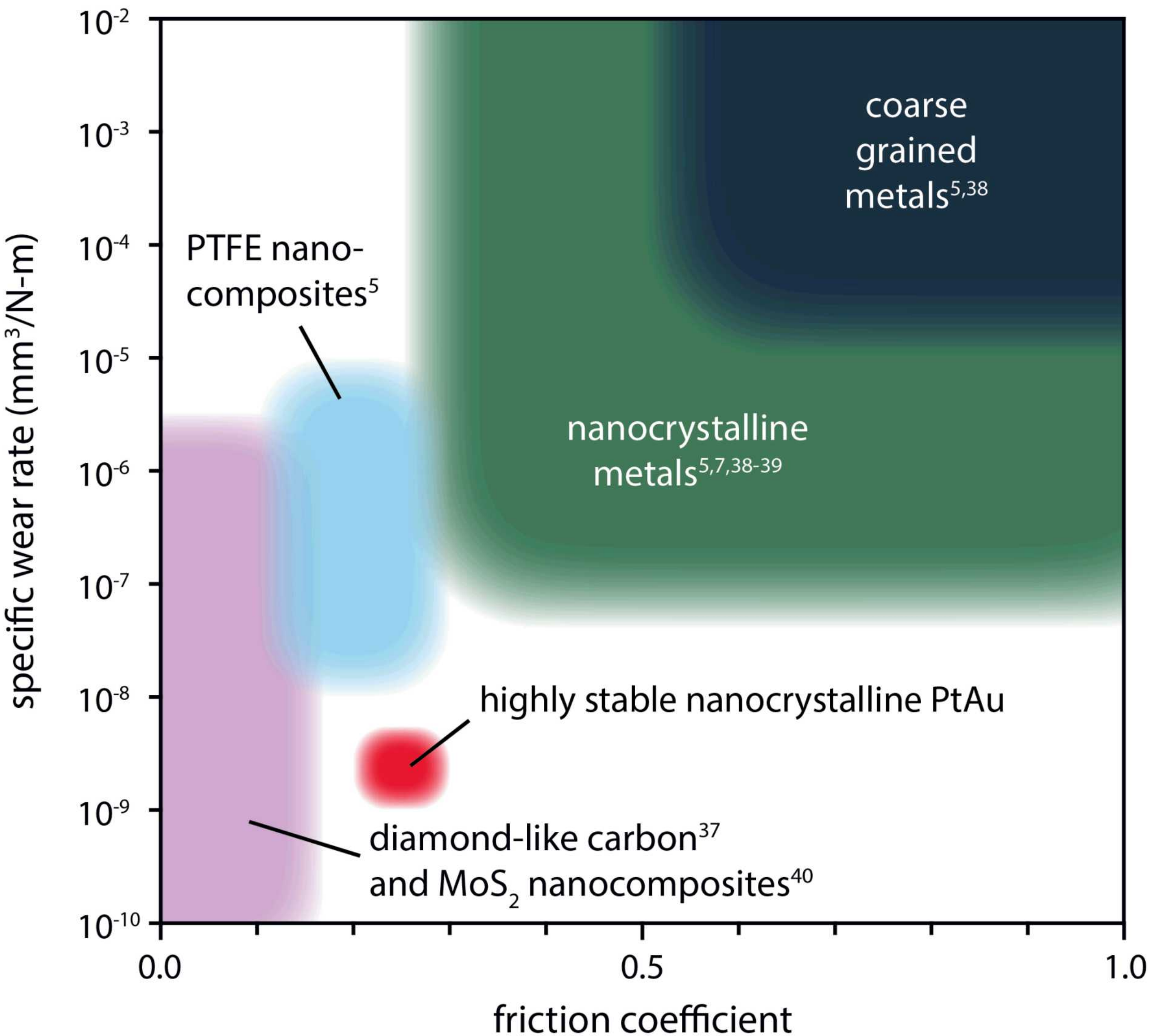


Alloying improves friction & wear performance by reducing and stabilizing grain size

...by reducing grain size:



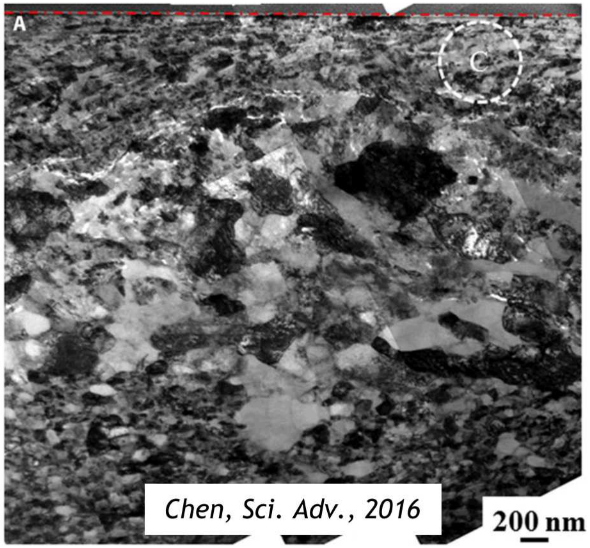
Low Friction and Wear of Nanocrystalline Metals



J. F. Curry, et al., ***Advanced Materials (2018)***

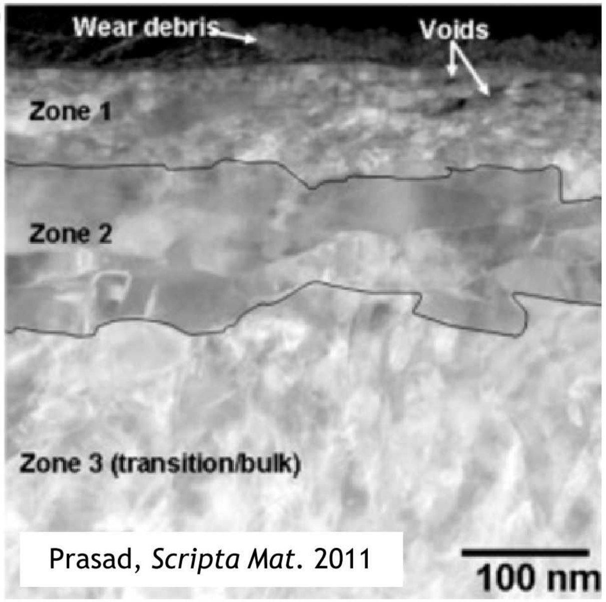
What do we know now?

Pure Cu in air @ 50 N, μ ~ 0.37



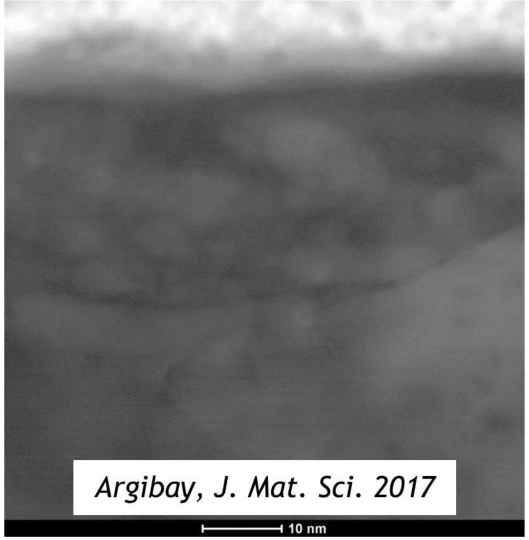
Chen, Sci. Adv., 2016

Pure Ni in dry N_2 @ 98 mN, μ ~ 0.2



Prasad, *Scripta Mat.* 2011

Pure Au in dry N_2 @ 1 mN, μ ~ 0.2

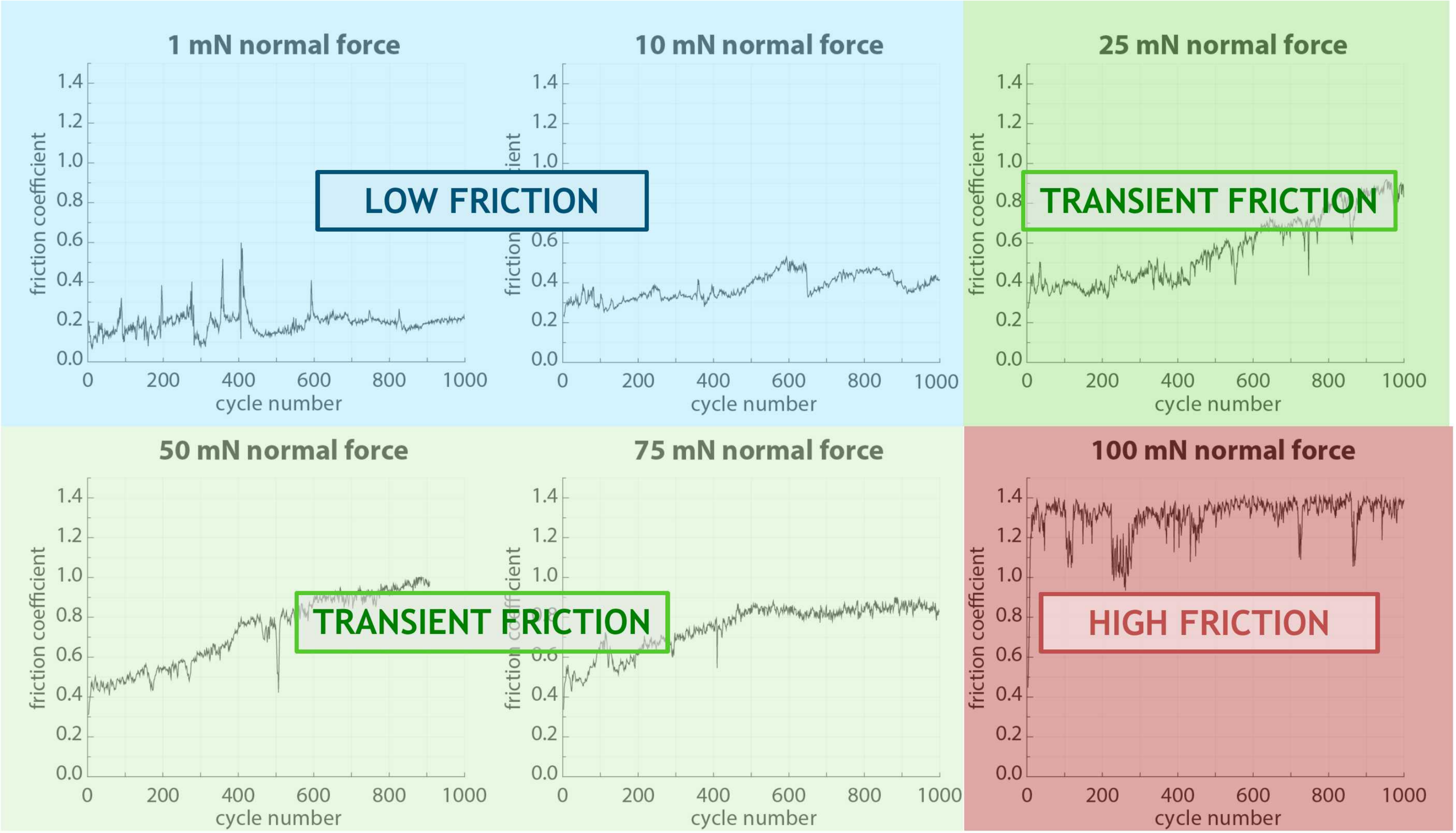


Argibay, J. Mat. Sci. 2017

Low friction is associated with the formation of a highly surface localized UNC layer.

What do we know? Friction depends on applied load

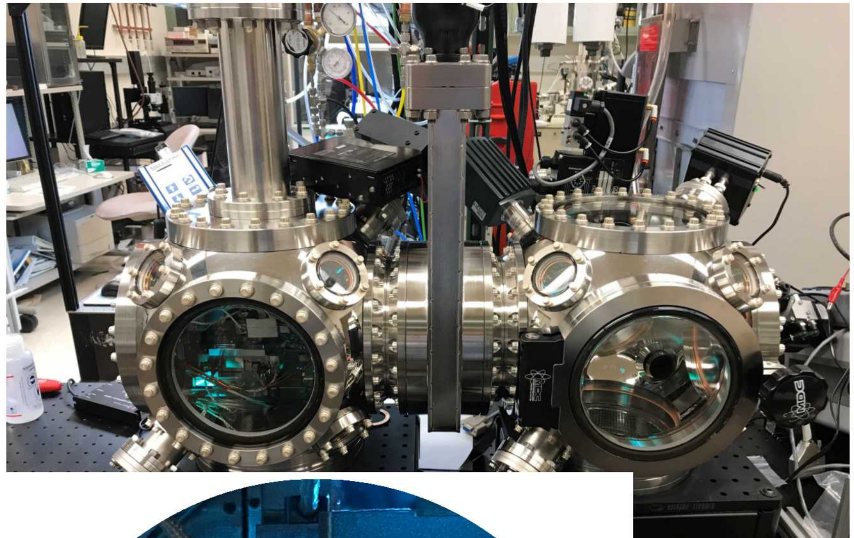
(Pure Au in dry N_2)



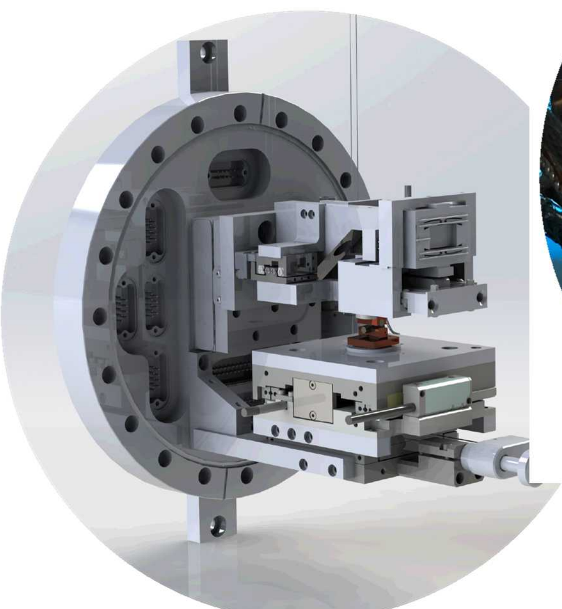
Three different friction regimes, with transitions.

What about adsorbates? Test in UHV

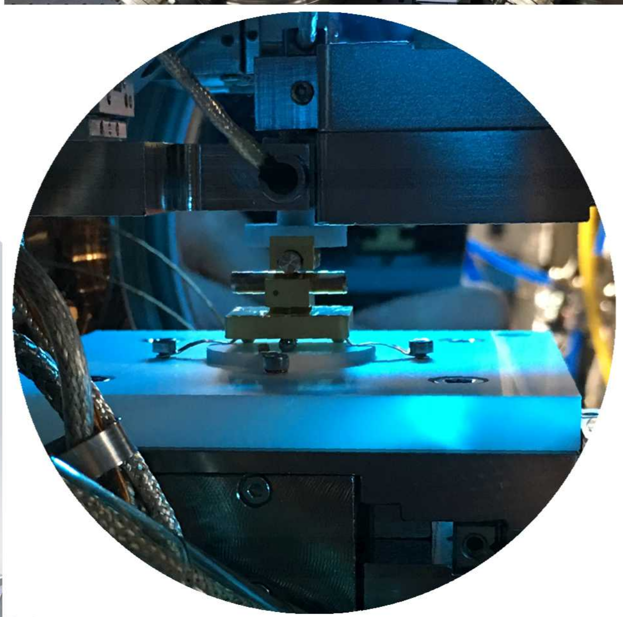
Left Module:
- Linear Reciprocator
- Load metering
- Cryo stage (4–800K)
- 10^{0} to 10^{-9} torr
- 0.1 mN to 1 N
- 100 μm/s – 100 mm/s
- 10 kHz acquisition
- capacitive displacement sensors & flexural cantilevers



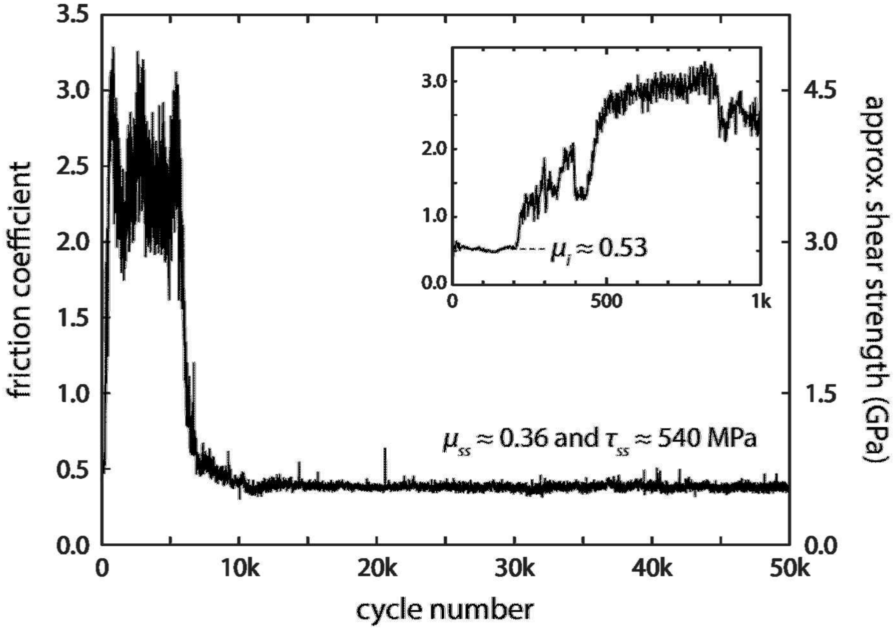
Right Module:
- Rotary Module
- Dead weight
- 5 mN to 10 N
- 100 Hz acquisition
- 10^{0} to 10^{-9} torr
- Strain gage sensor

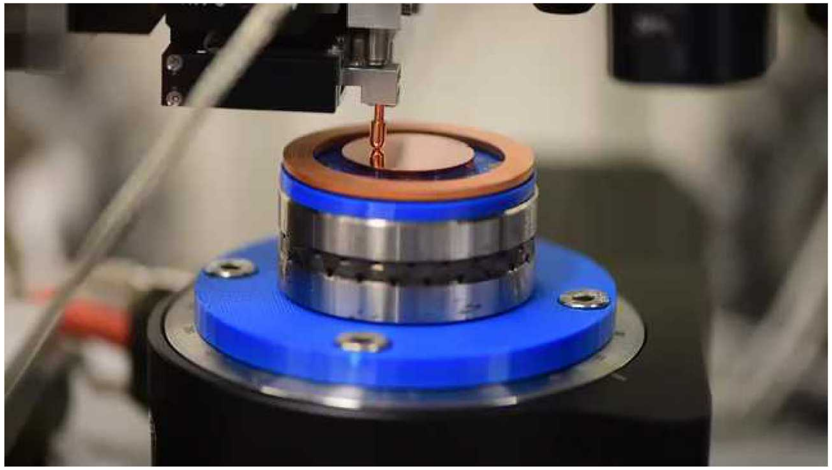


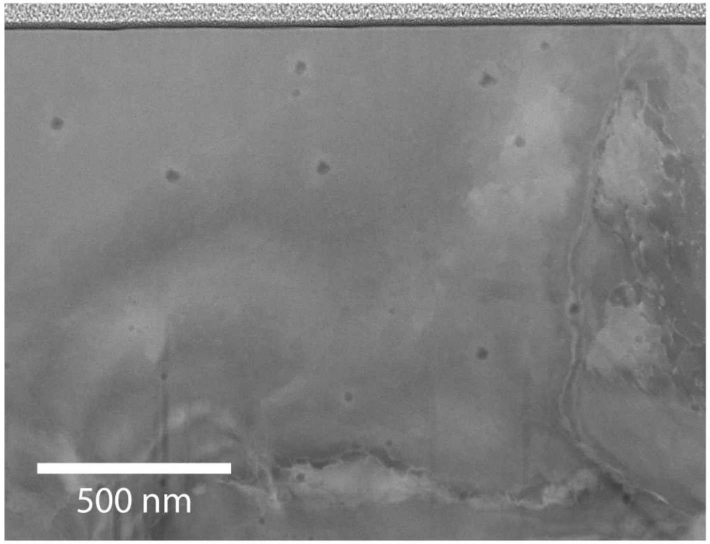
crossed-cylinders and sphere-on-flat configurations



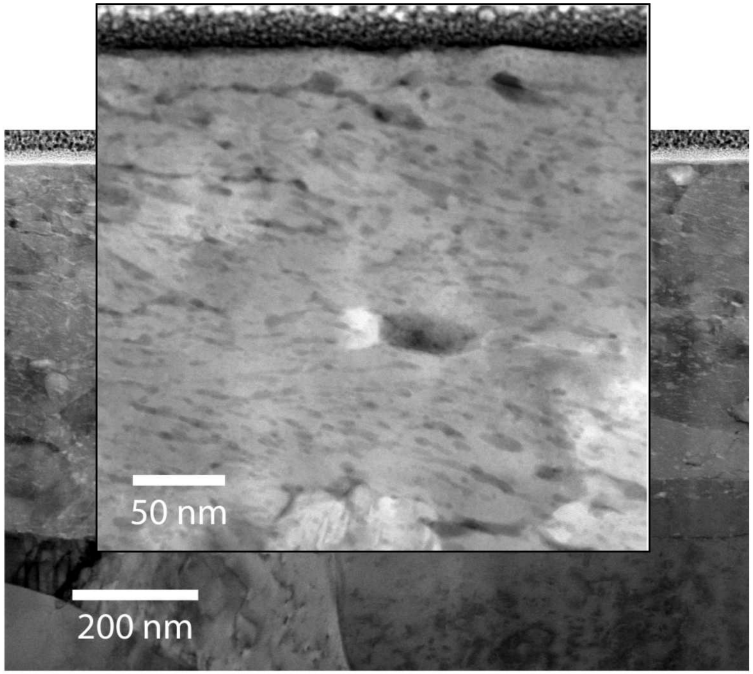
Low friction of FCC metals (pure Cu) in ultra-high vacuum



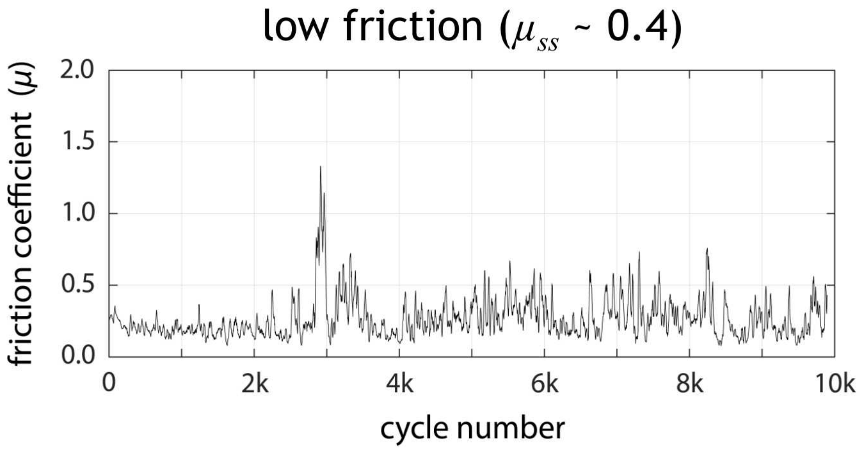


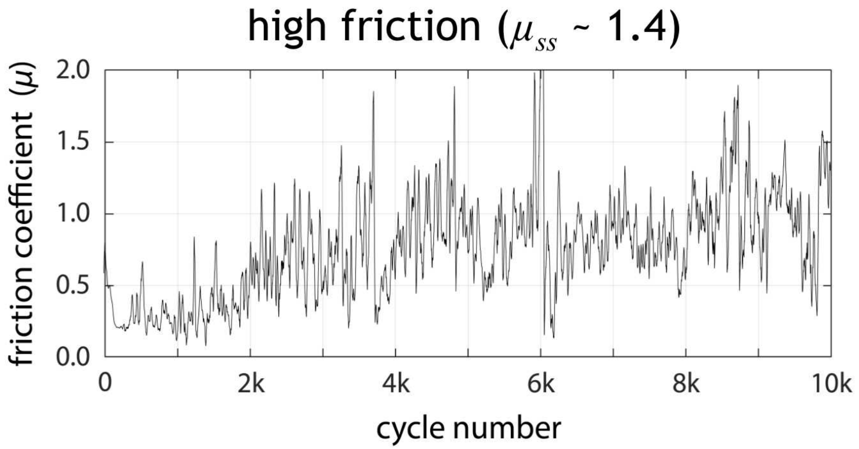


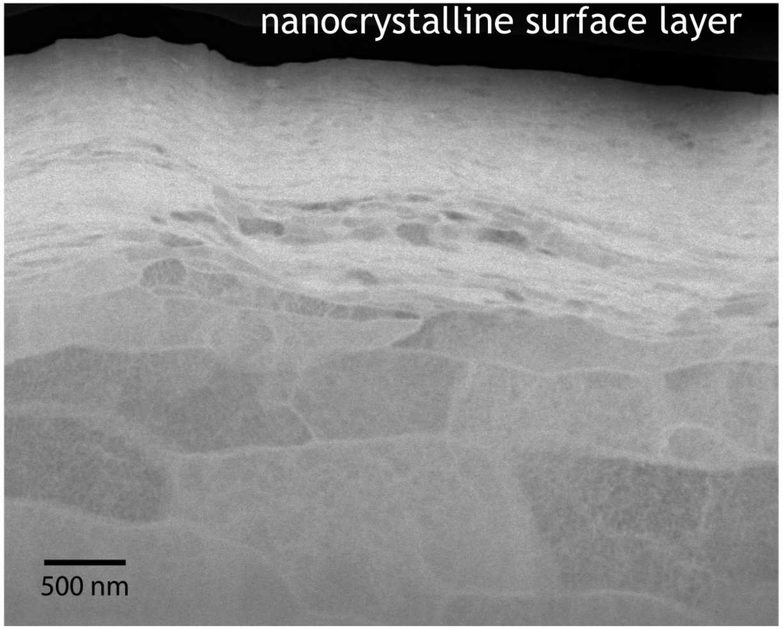
refinement

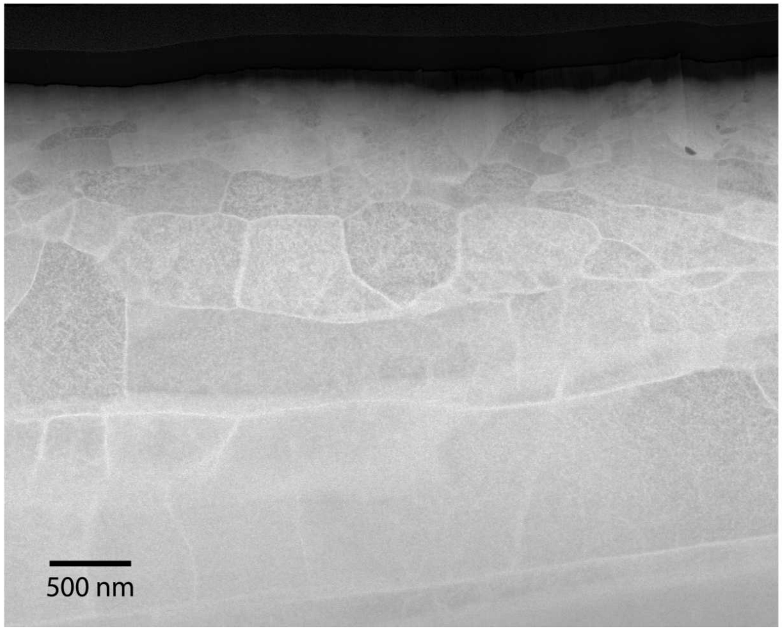


Low friction of BCC metals (pure Ta) in ultra-high vacuum

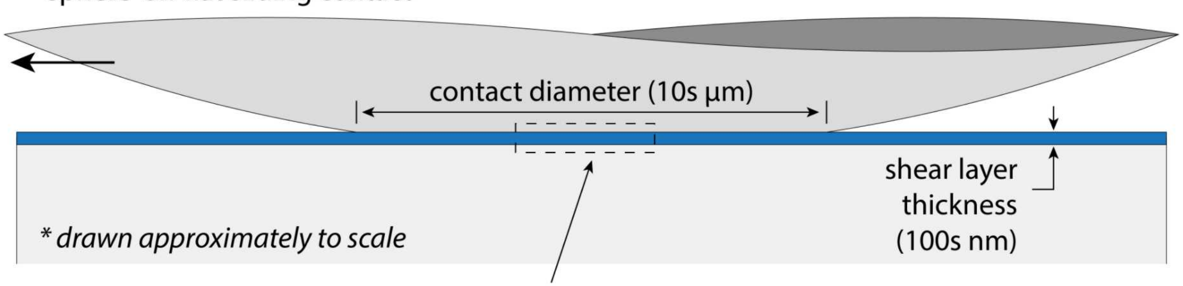


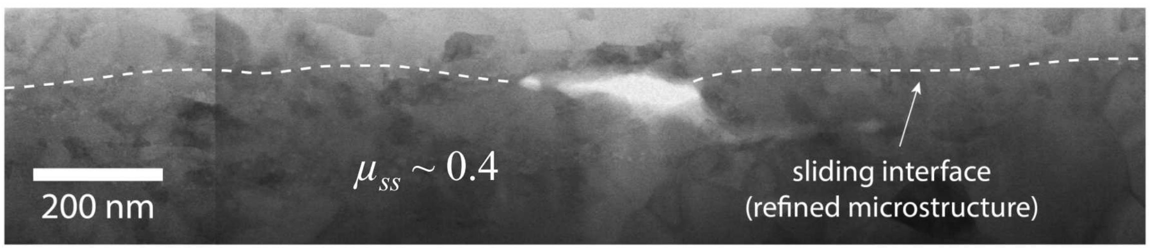




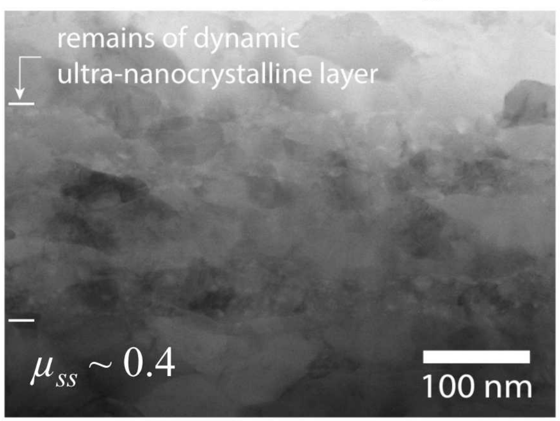


A cross-sectional view of metal sliding contacts

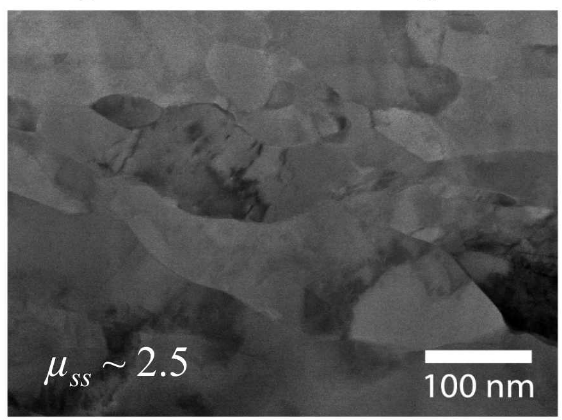




low friction and ultra-fine grains

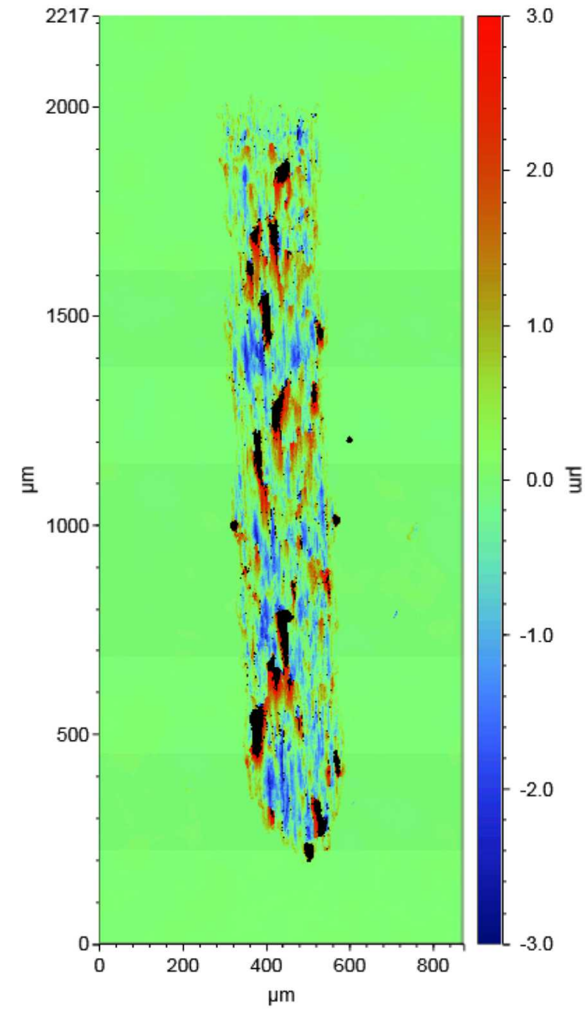


high friction and coarser grains



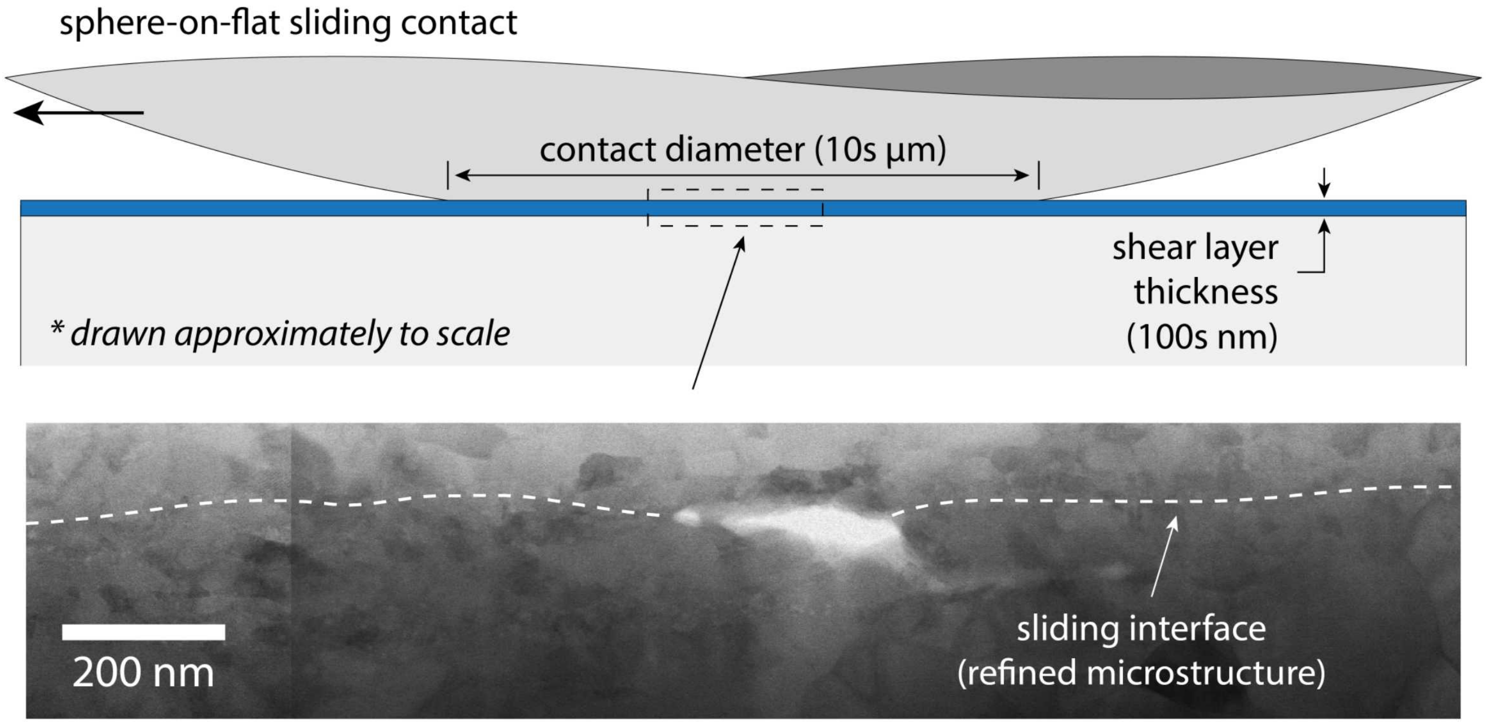
(TEM of self-mated pure Cu in dry N_2)

wear track topography map:



Contact diameters ~ 10s μm
Shear layers ~ 10s nm thick

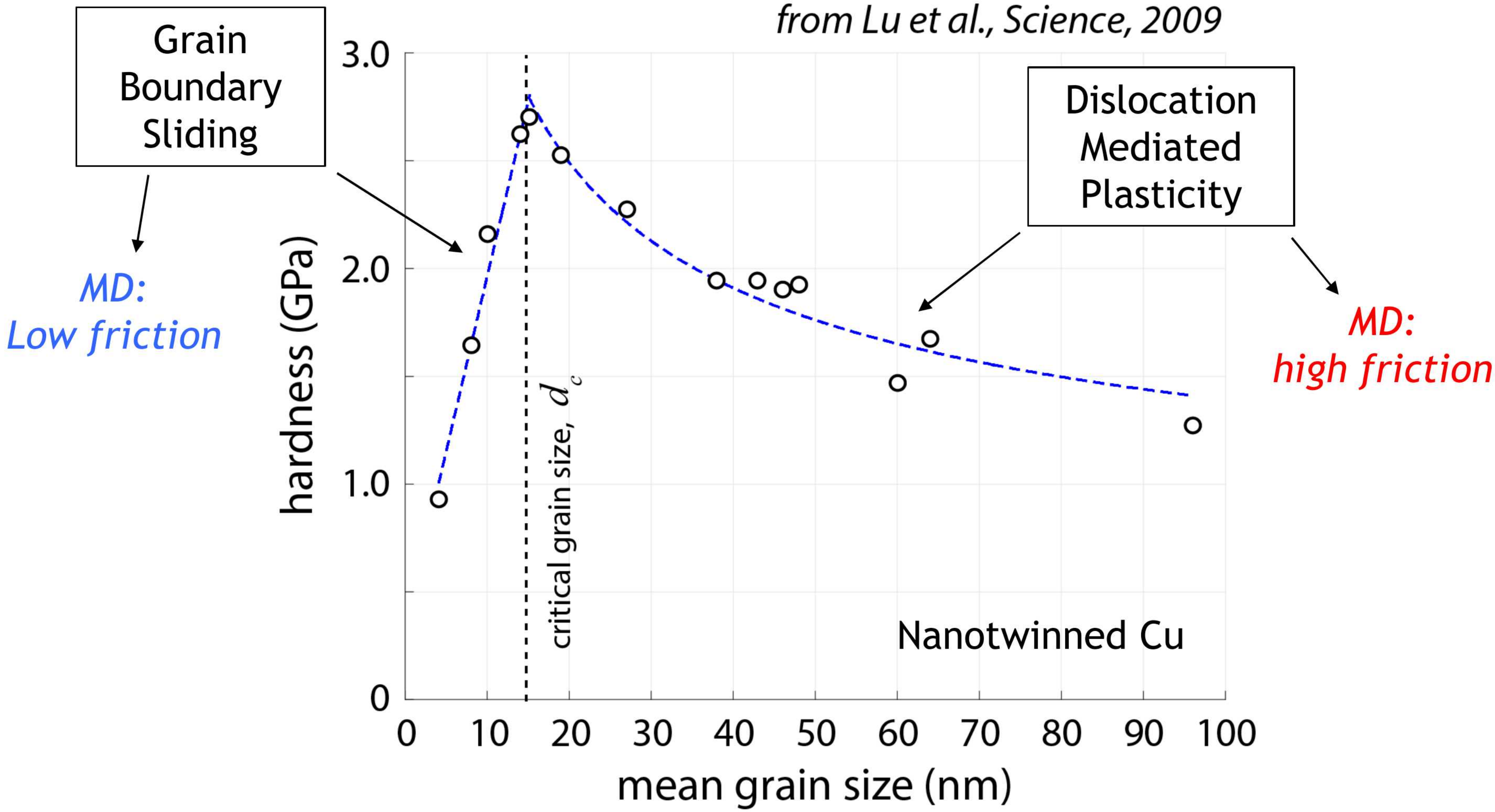
The basis of friction in metals



For metals, the friction coefficient can be described as the ratio of interface shear strength and bulk hardness:

$$\mu = \frac{\tau}{H}$$

How does grain size affect materials properties?

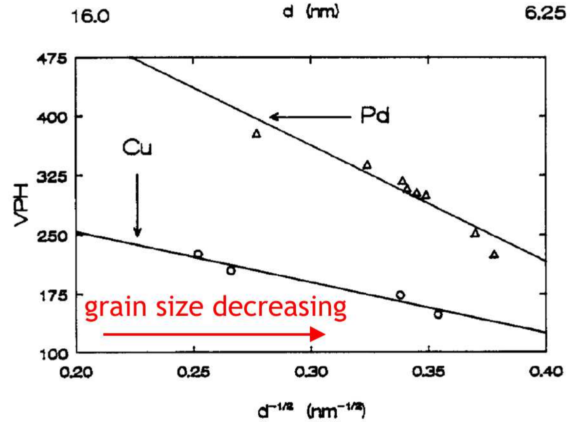


Details of MD simulations can be found in Argibay, et al., ***J. Mat. Sci. (2017)***

Hall-Petch breakdown occurs at about 10 nm (critical grain size)

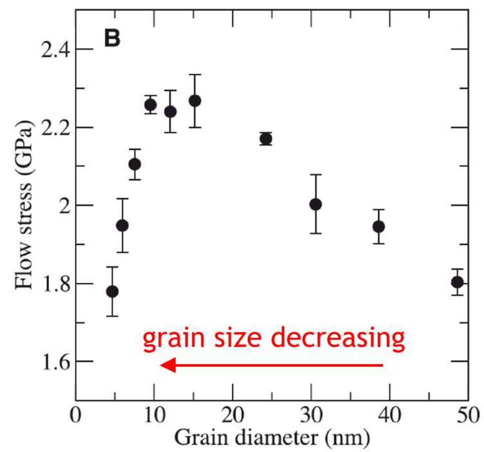
FCC Cu and Pd experiments

Chokshi, et al., Scripta Mat., 1989



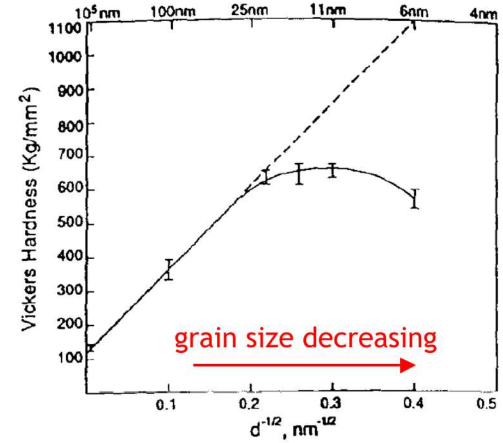
FCC Cu simulations

Schiötz and Jacobsen, Science, 2003



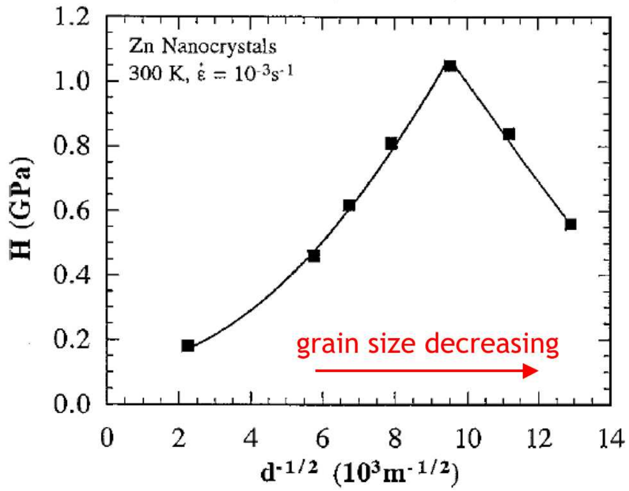
FCC Ni experiments

Erb, Nanostructured Matls., 1995



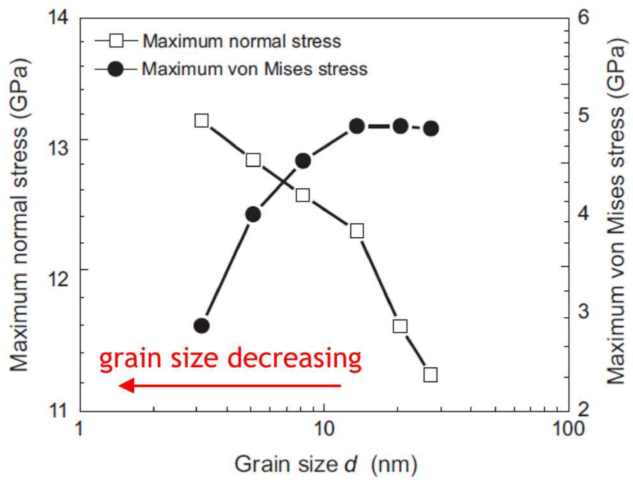
HCP Zn experiments

Conrad and Narayan, App. Phys. Lett., 2002



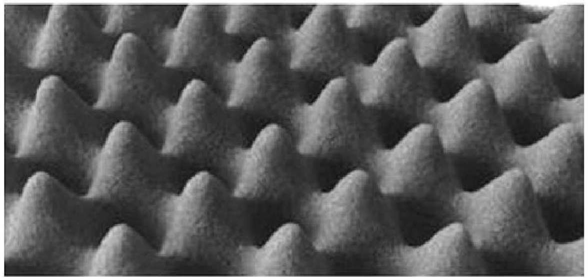
BCC Ta simulations

Tang, et al., Mat. Sci. & Eng. A, 2013

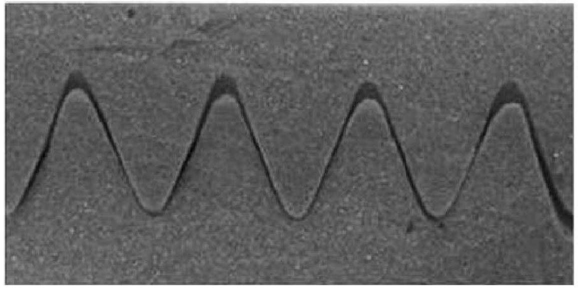


The Mechanism Behind Low and High Friction in Metals

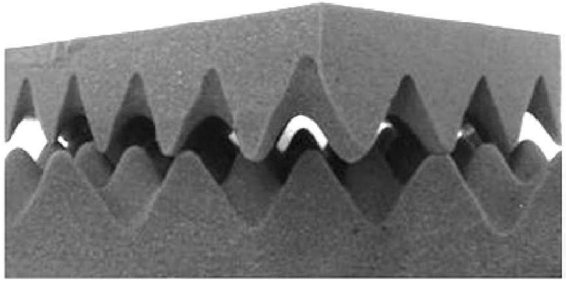
Atomic lattices have periodicity



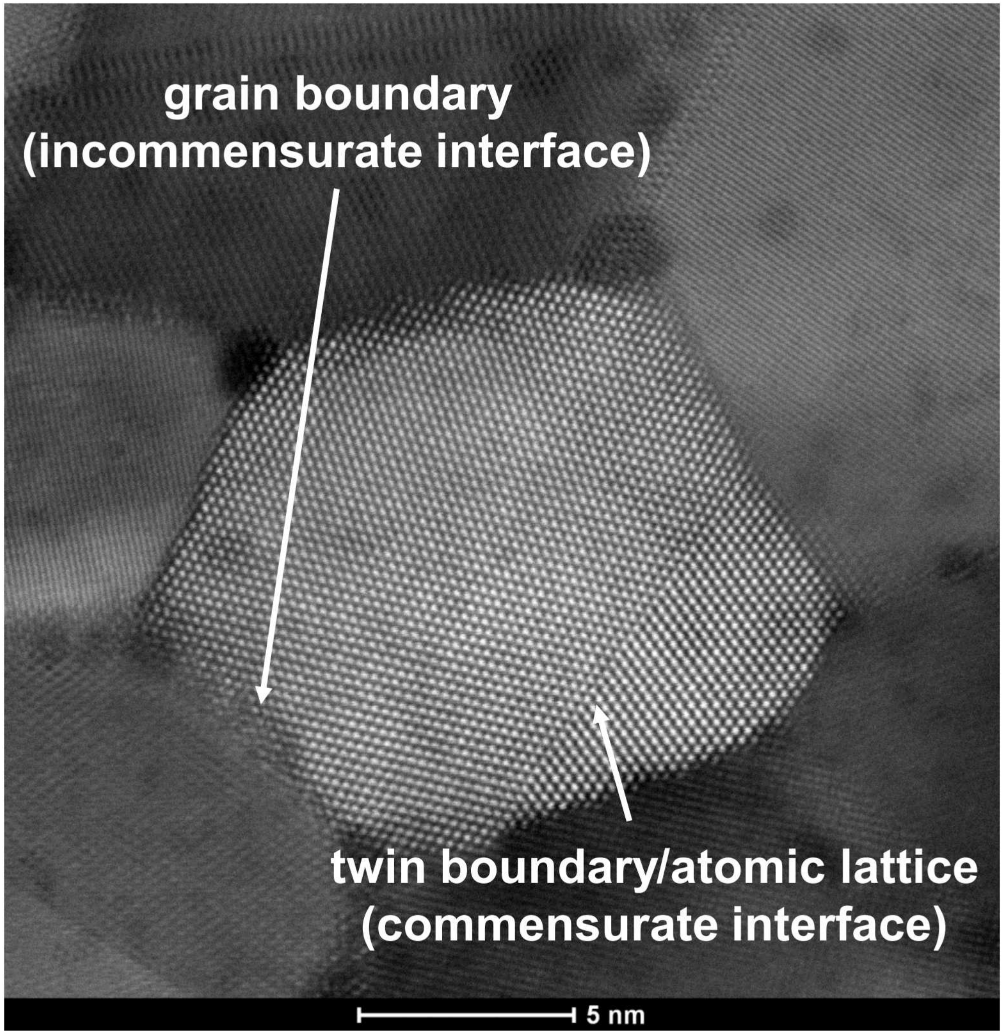
Commensurate interface
(higher energy barrier/higher μ)



Incommensurate interface
(lower energy barrier/low μ)



High resolution TEM



Evidence of Intragranular Amorphization

From Kaibyshev, Mat. Sci. & Eng., 2002:

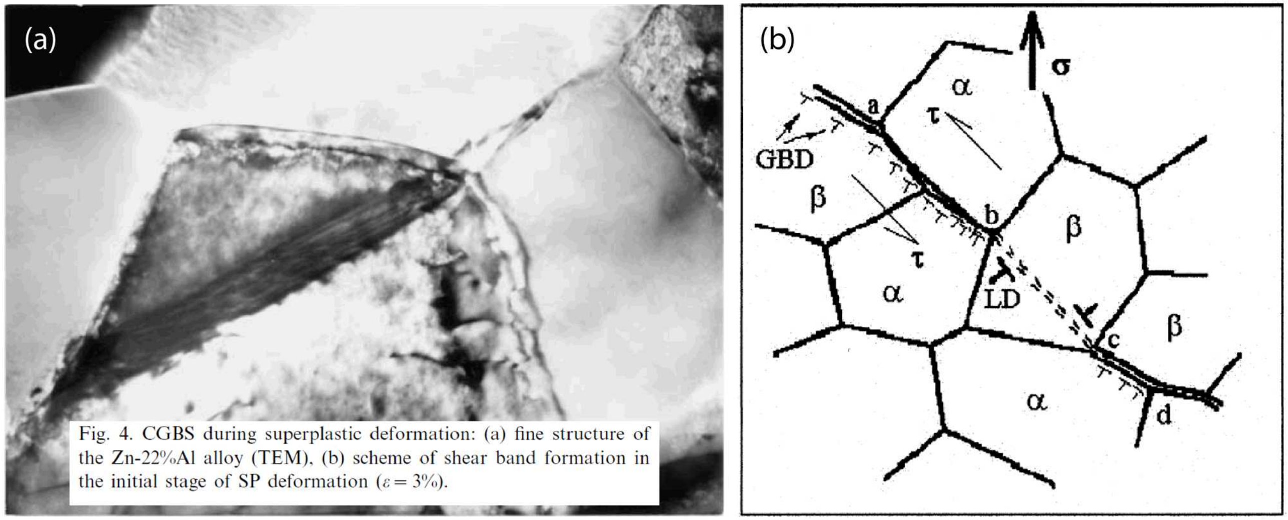
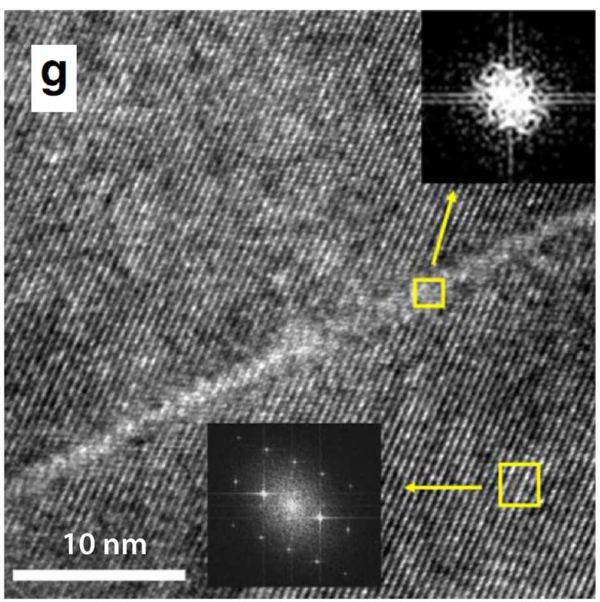
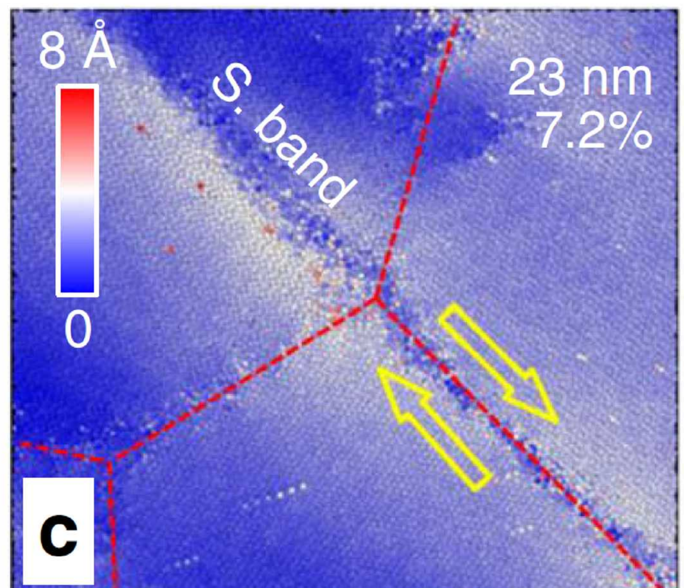


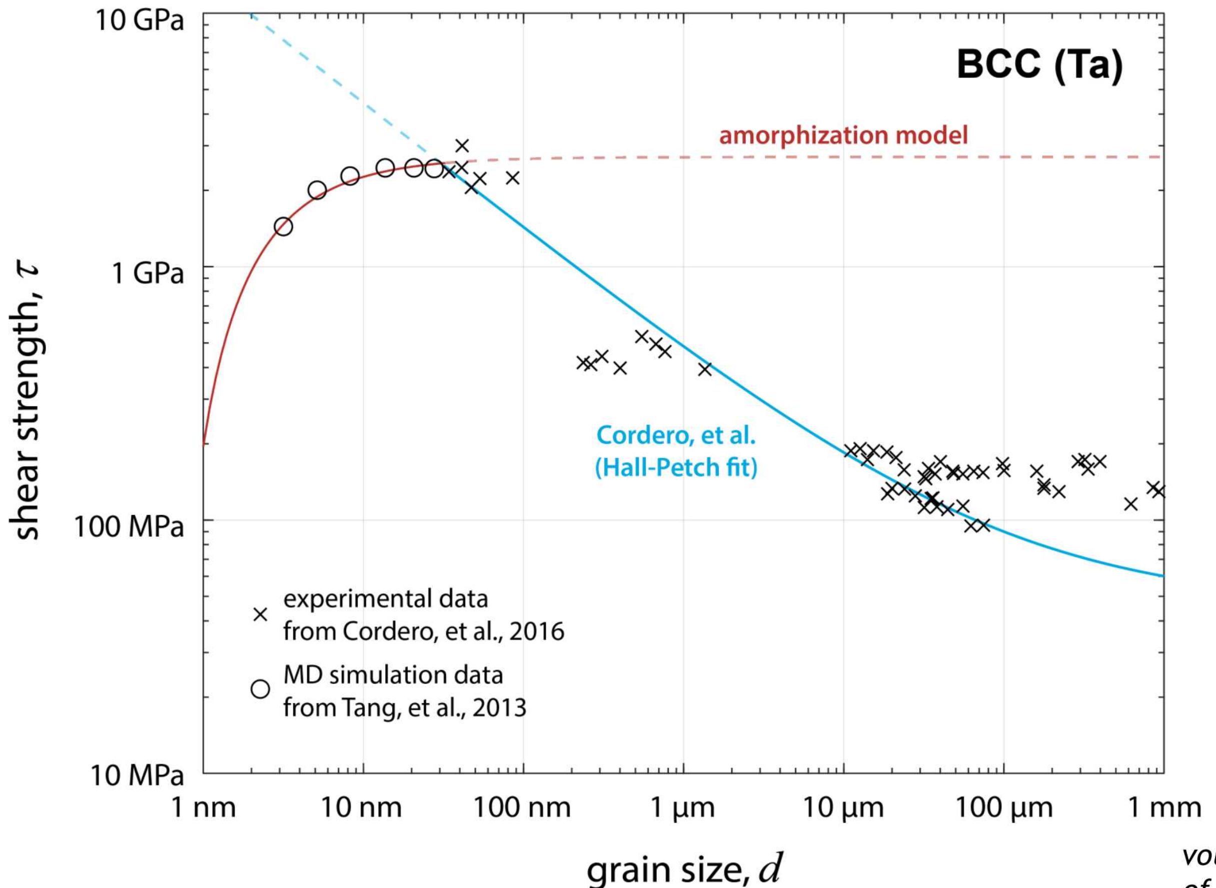
Fig. 4. CGBS during superplastic deformation: (a) fine structure of the Zn-22%Al alloy (TEM), (b) scheme of shear band formation in the initial stage of SP deformation ($\varepsilon = 3\%$).

From Luo, et al., Nat. Comm., 2019:





Grain Size Dependent Strength of Metals



Amorphization model:

Chandross and Argibay (in peer review)

$$\tau(d) = L\left(1 - \frac{T}{T_m}\right)\frac{\rho_L}{M}\left(\frac{d-\delta}{d}\right)^3 - \frac{kT}{b^3}$$

volume fraction of GB to crystal

heat of fusion

homologous temperature

amorphous metallic atomic volume

thermal energy

Back to the future… an improved interpretation of old concepts

Rosenhain and Ewen predict amorphous g

THE INTERCRYSTALLINE
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By WALTER ROSENHAIN,

DONA

(Both of the Nat

In their first paper on t
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in favour of an hypothesis
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be briefly termed the " am
that the crystals of which
" cemented " together by
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different physical state.
intercrystalline layer is reg
least closely analogous to
undercooled liquid which
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meet one another in variou

Slip at Grain Boundaries and Grain Growth in Metals

By N. F. MOTT,
H. H. Wills Physical Laboratory, Bristol

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(*b*) The observed fact that at the melting point the slip is the same as that which would be given by a monomolecular layer of liquid appears in the theory as an *accident*. The mechanism of flow in liquid aluminium cannot be anything like that sketched here, because the temperature dependence is 10–20 times smaller; the viscosity of liquid metals depends on temperature according to the formula

$$\sigma = \sigma_0 e^{W/kT},$$

where W is of the order of the latent heat of fusion (Frenkel 1946).

We must therefore modify our hypothesis. Let us suppose that the elementary act which allows slip to occur is the *disordering* of atoms round each island where fit is good. The free energy F necessary to do this will approach zero at the melting point and nL at the absolute zero of temperature; here L is the latent heat of fusion per atom. At any other temperature, let us assume F to be given by

$$F = nL(1 - T/T_M),$$

where T_M is the temperature of melting. Let us assume also, since the disordering will result in a slip through a distance a, that a stress σ will decrease or increase F by $\pm\frac{1}{2}\sigma n\omega a$. Then the rate of slip is now

$$v = 2\nu a \exp\{-nL(1 - T/T_M)/\boldsymbol{k}T\}\sinh(\sigma n\omega a/2\boldsymbol{k}T),$$

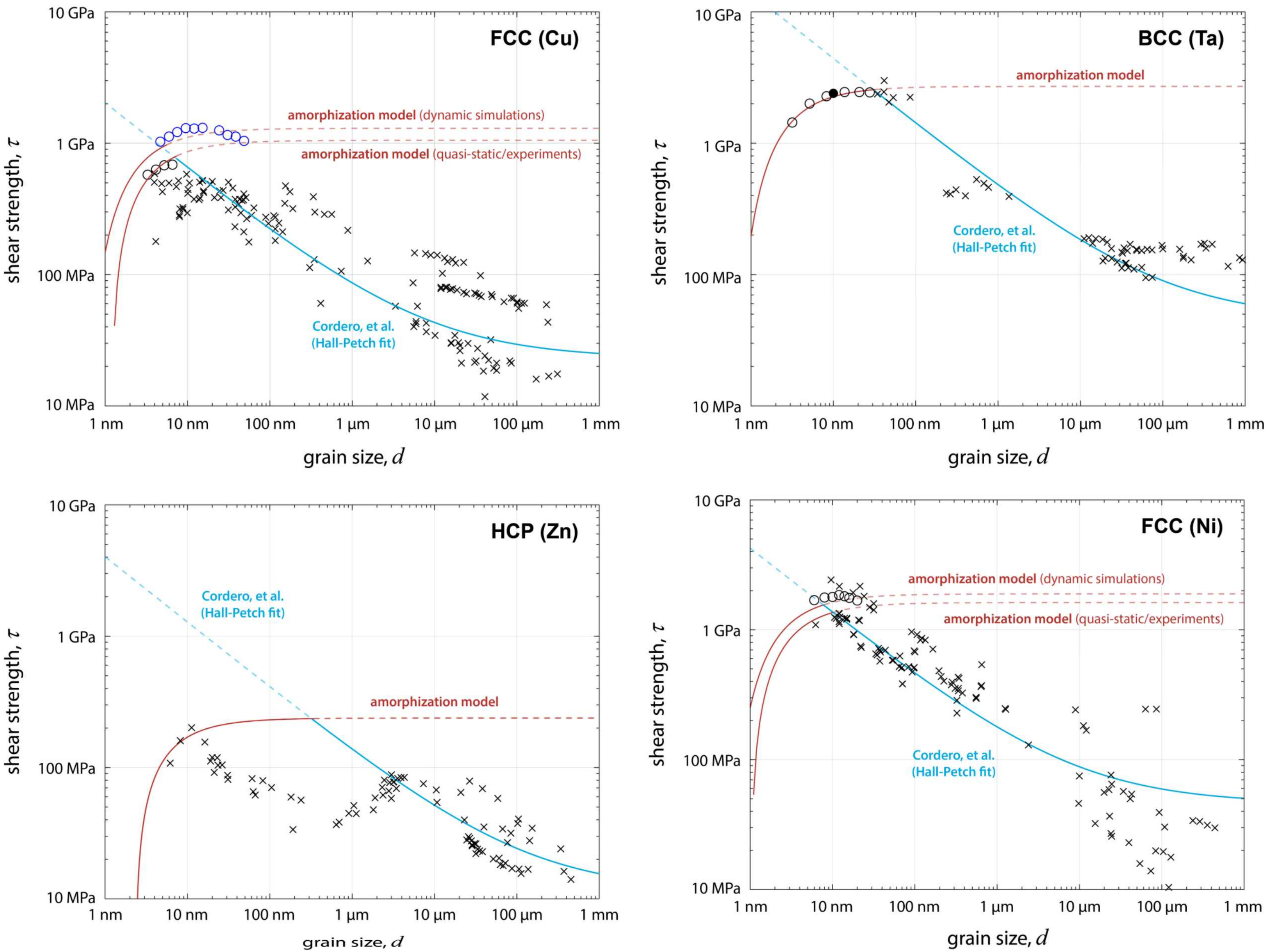
which for small σ reduces to

$$v = \frac{\nu a^2 n\omega\sigma}{\boldsymbol{k}T}\exp\left(\frac{nL}{\boldsymbol{k}T_M}\right)\exp\left(\frac{-nL}{\boldsymbol{k}T}\right).$$

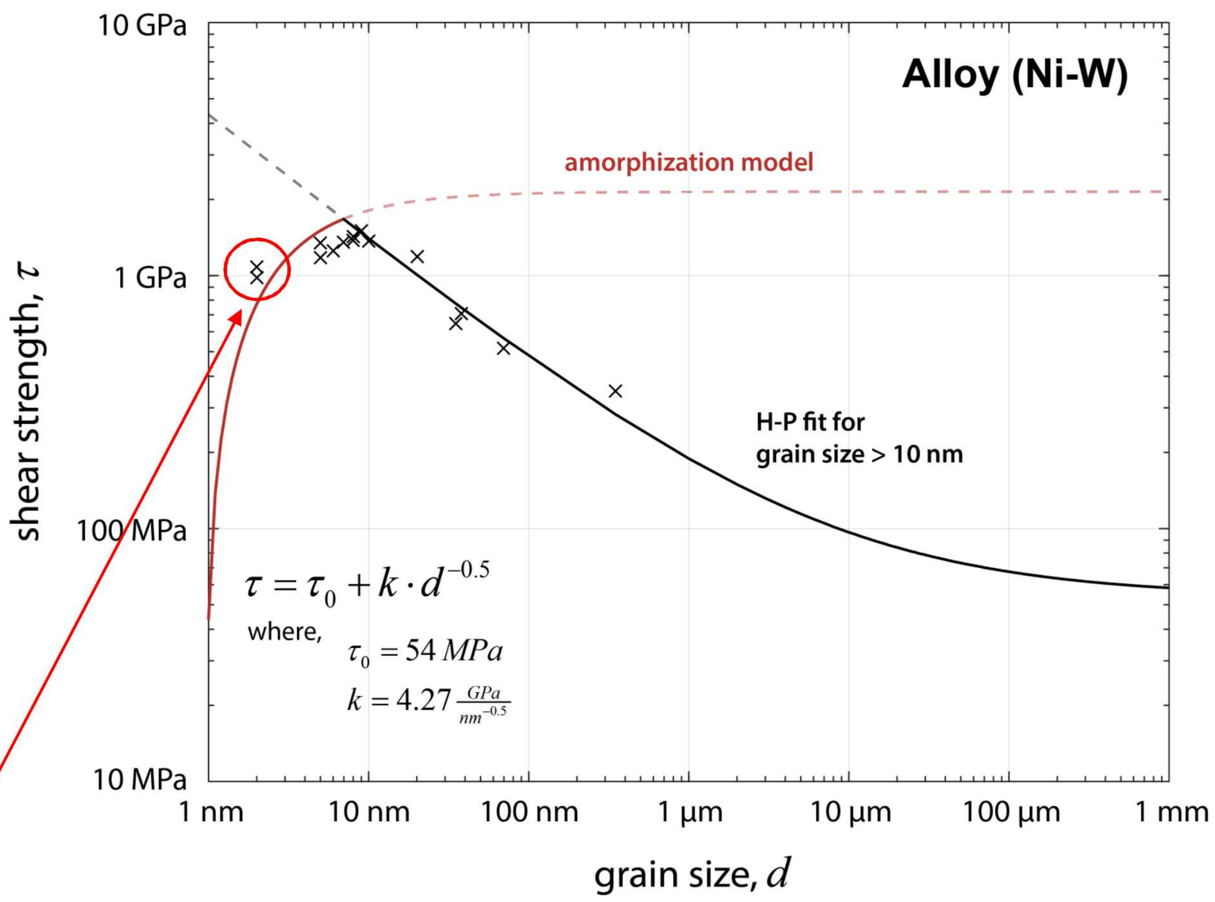
Mott provided a theory of slip based on the formation of disordered, liquid-like islands of atoms

A review of these concepts is currently under peer-review at Applied Physics Reviews

The model works well for many other metals



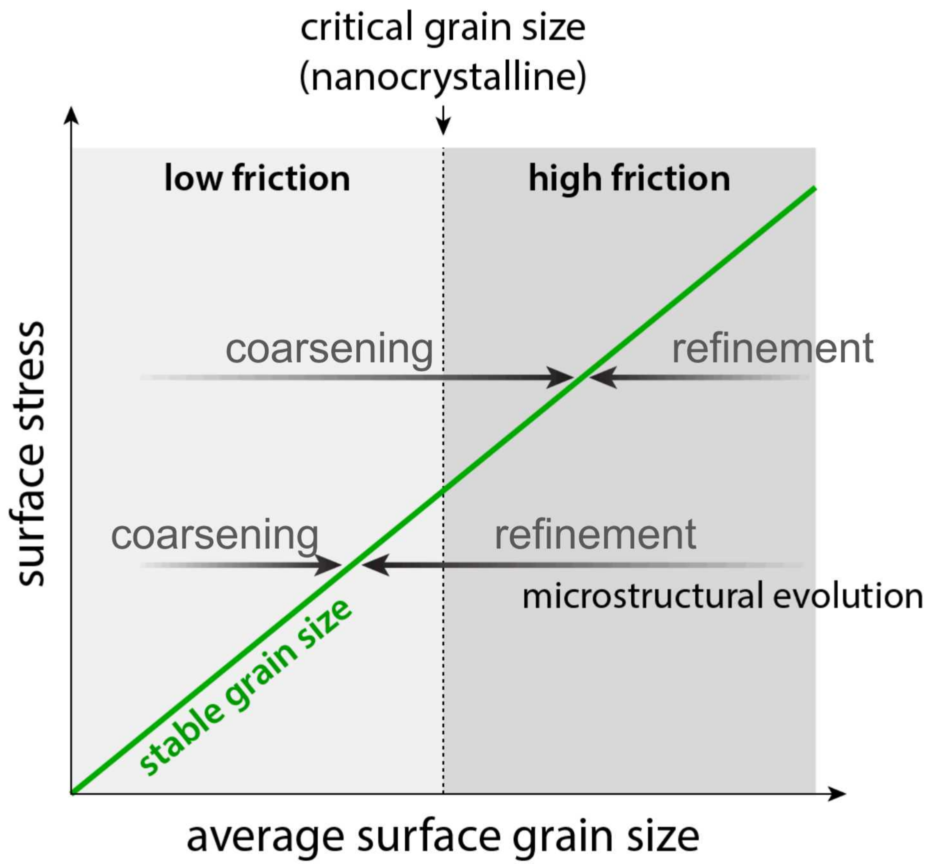
Amorphization model also works for alloys

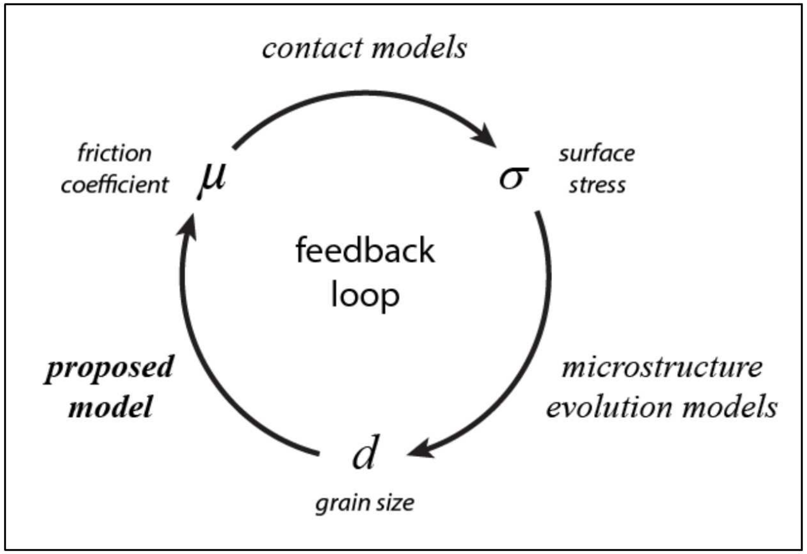


Keep in mind:

1. Grain size estimates are particularly difficult and exhibit higher uncertainty in the 1-10 nm range, and
2. these errors are relatively small compared to typical H-P fit for far coarser grain size (e.g. earlier pure metal data)

Proposed stress-dependent steady-state (asymptotic) grain size

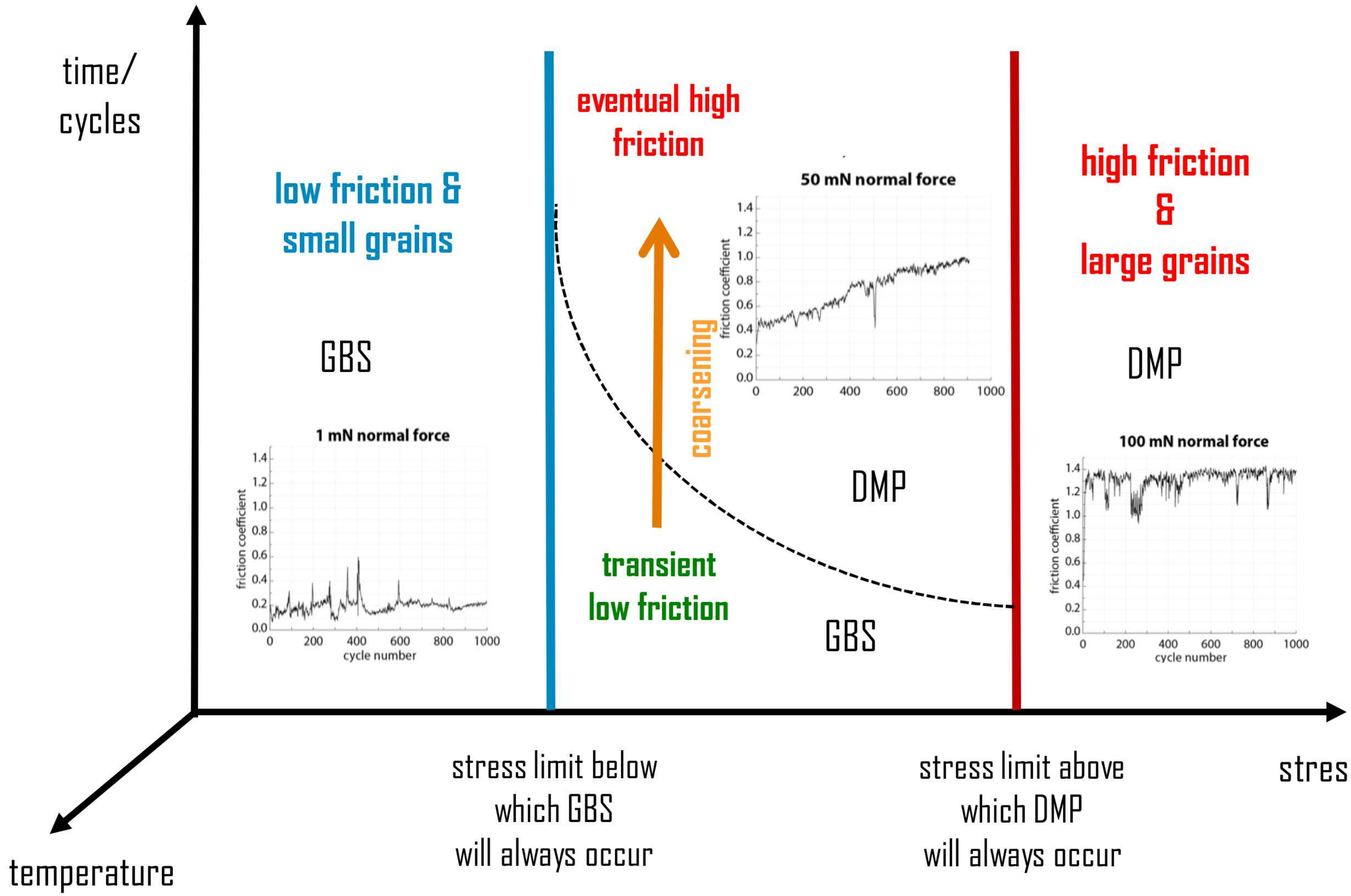




- Effective refinement from recrystallization (Zener & Holloman, 1944; McQueen et al., 1967)
- Known in rocks and ice cores (Derby et al., 1992)
- Recently extended to metals under severe plastic deformation (Pougis et al., 2014)

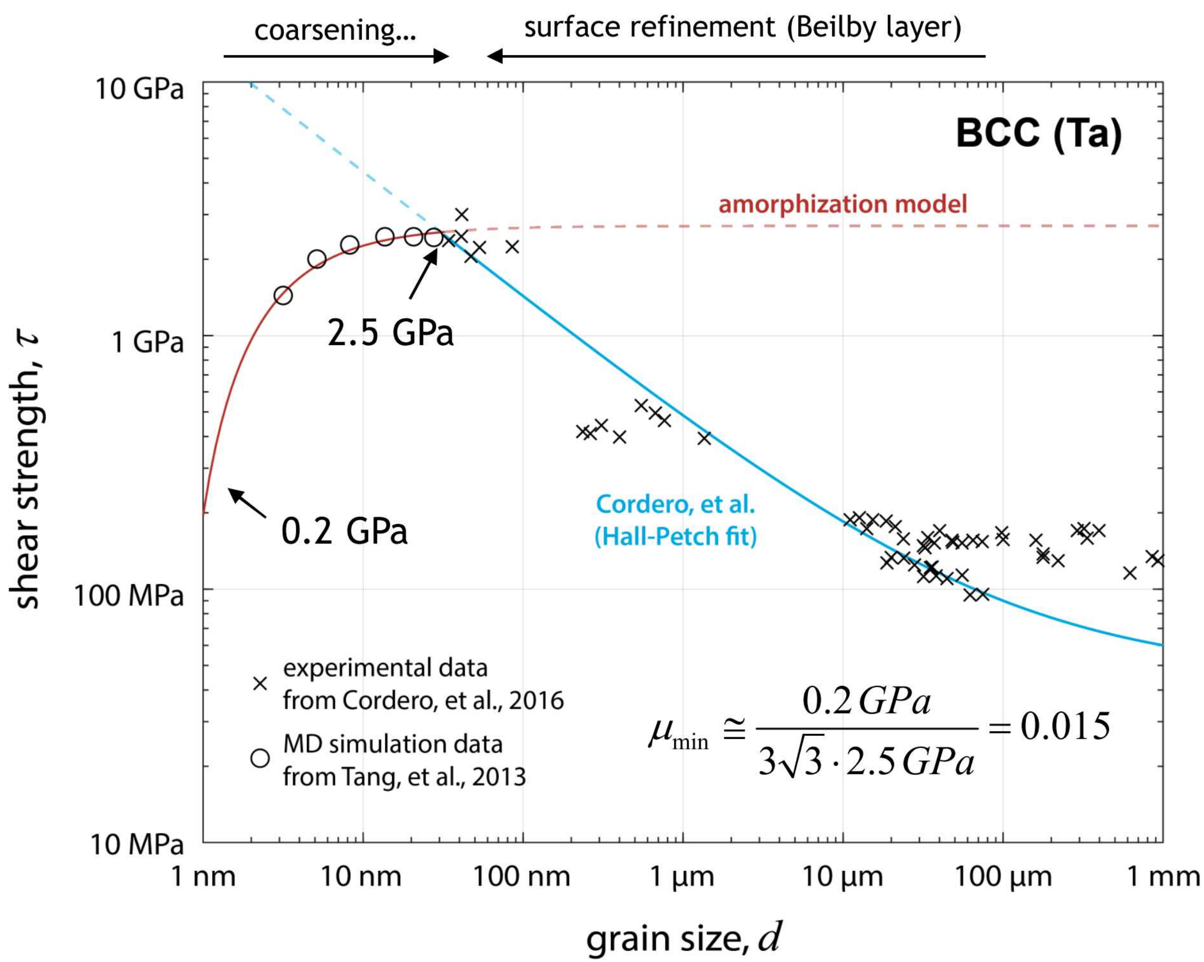
Argibay, et al., ***J. Mat. Sci. (2017)***

Generalized friction regimes map for metals



Argibay, et al., **J. Mat. Sci. (2017)**

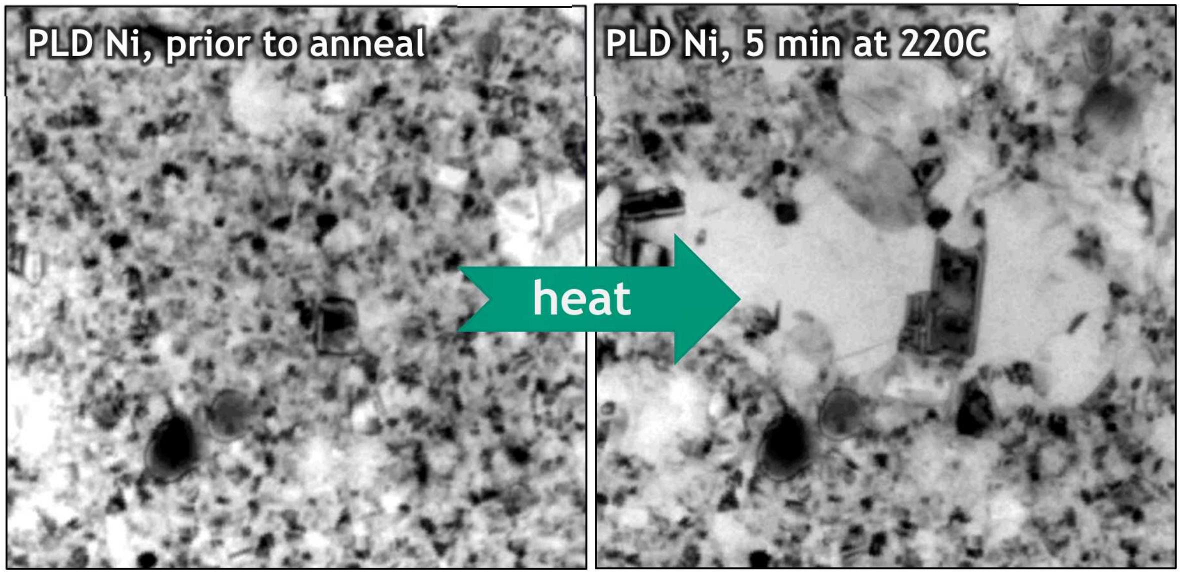
What is the lowest friction condition?



$$\mu_{\min} \cong \frac{0.2\,GPa}{3\sqrt{3}\cdot 2.5\,GPa} = 0.015$$

Bowden & Tabor: $\mu = \frac{\tau}{H}$ $\xrightarrow{\text{Tabor}}$ $\mu = \frac{\tau}{3\cdot\sigma_Y}$ $\xrightarrow{\text{Von Mises}}$ $\mu = \frac{\tau(d_{surface})}{3\sqrt{3}\cdot\tau(d_{bulk})}$

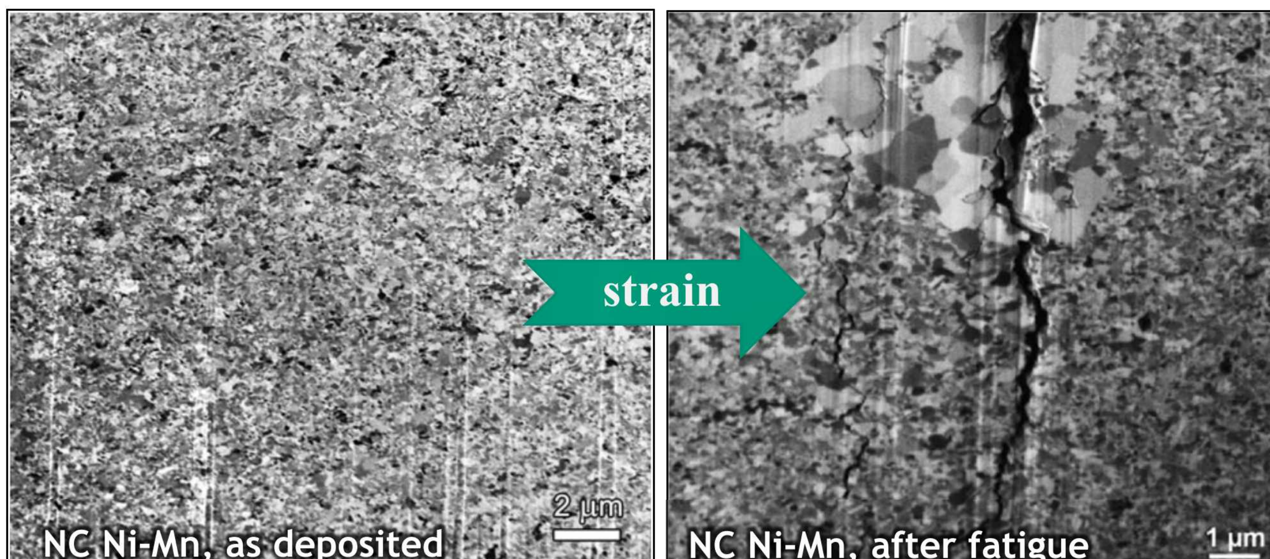
The challenge of grain size instability…



Grain growth in nanocrystalline metals is driven

Thermally…

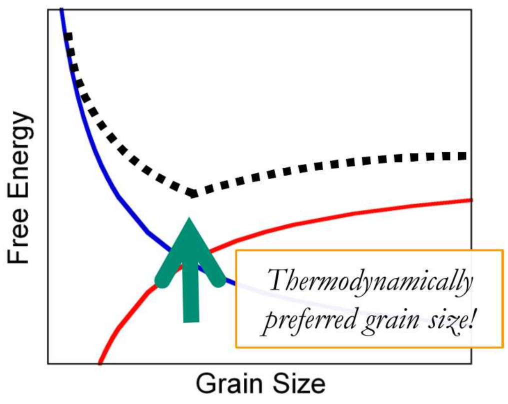
…and Mechanically



S. Rajasekhara, K. Hattar, P. Ferreira, A. Kinghorn, B. G. Clark, unpublished.;H. A. Padilla and B. L. Boyce, Exp. Mech. (2010).

Regular Nanocrystalline Solution Model (Chris Schuh Group at MIT)

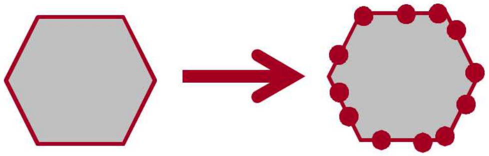
Slide courtesy of C. Schuh and H. Murdoch (MIT)



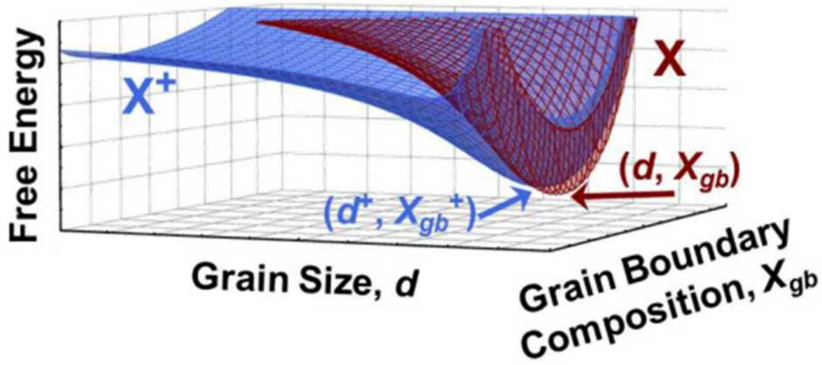
RNS Model:

Binary metal alloys exist possessing highly (intrinsically?) thermodynamically stable nanocrystallinity.

Solute segregation at grain boundaries:

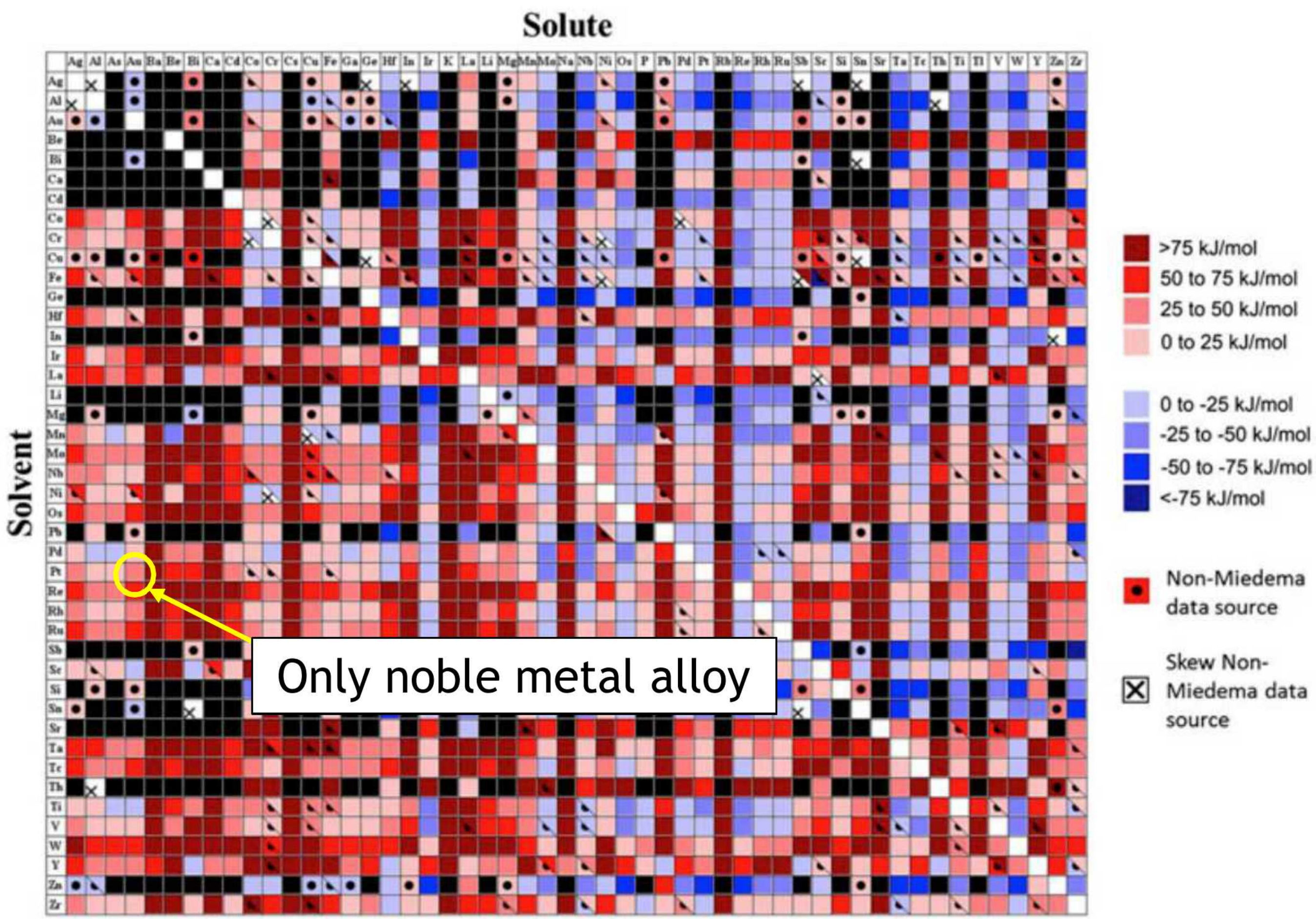


$$dG = \left[\gamma - \frac{N_\beta}{A} \Delta G_{seg} \right] dA$$



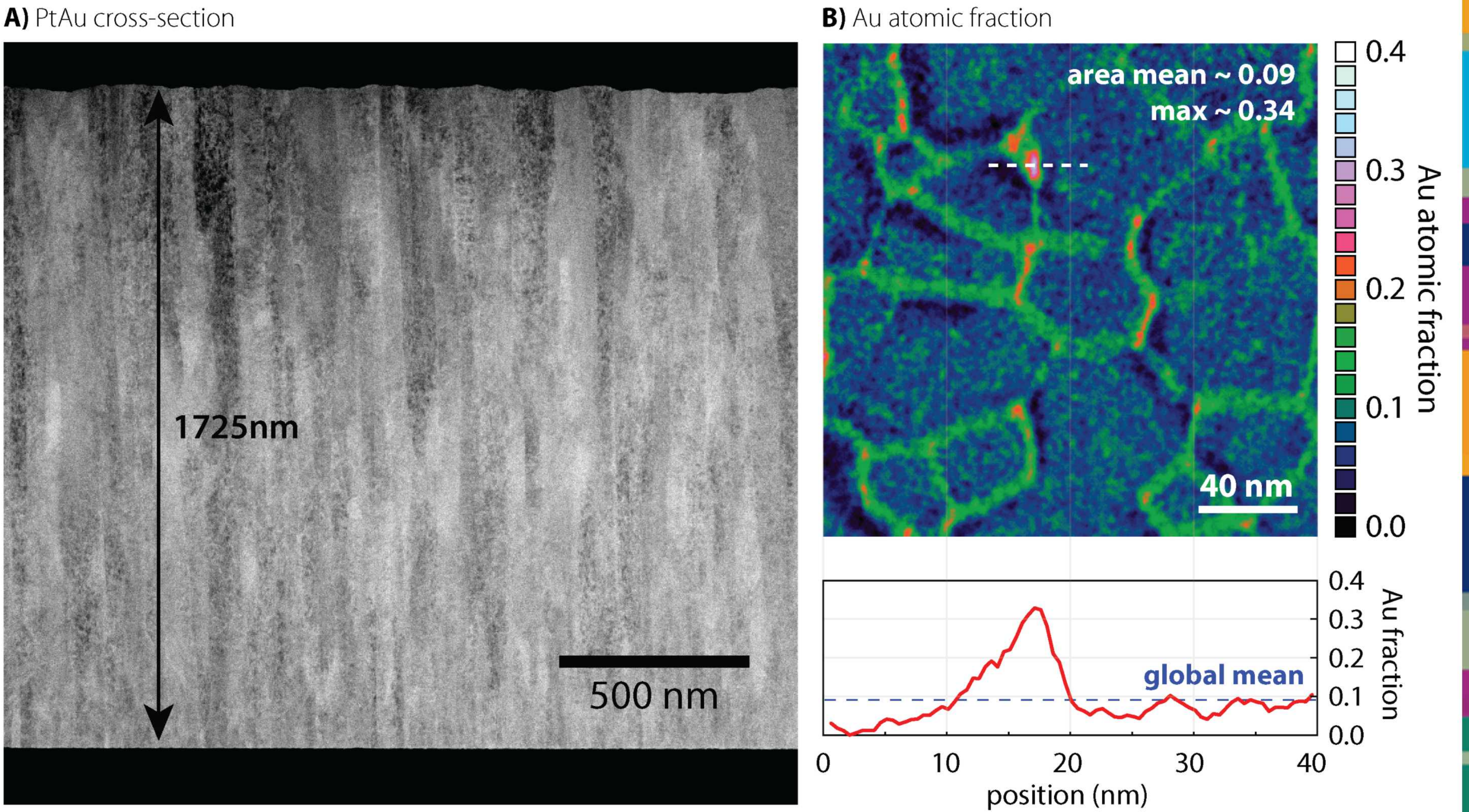
References:
T. Chookajorn, et al., Science, 2012
Kirchheim, Acta Materialia, 2002
Weissmuller, J. Materials Research, 1994
D.S. Gianola et al., Acta Materialia, 2006

So many alloys to choose from!



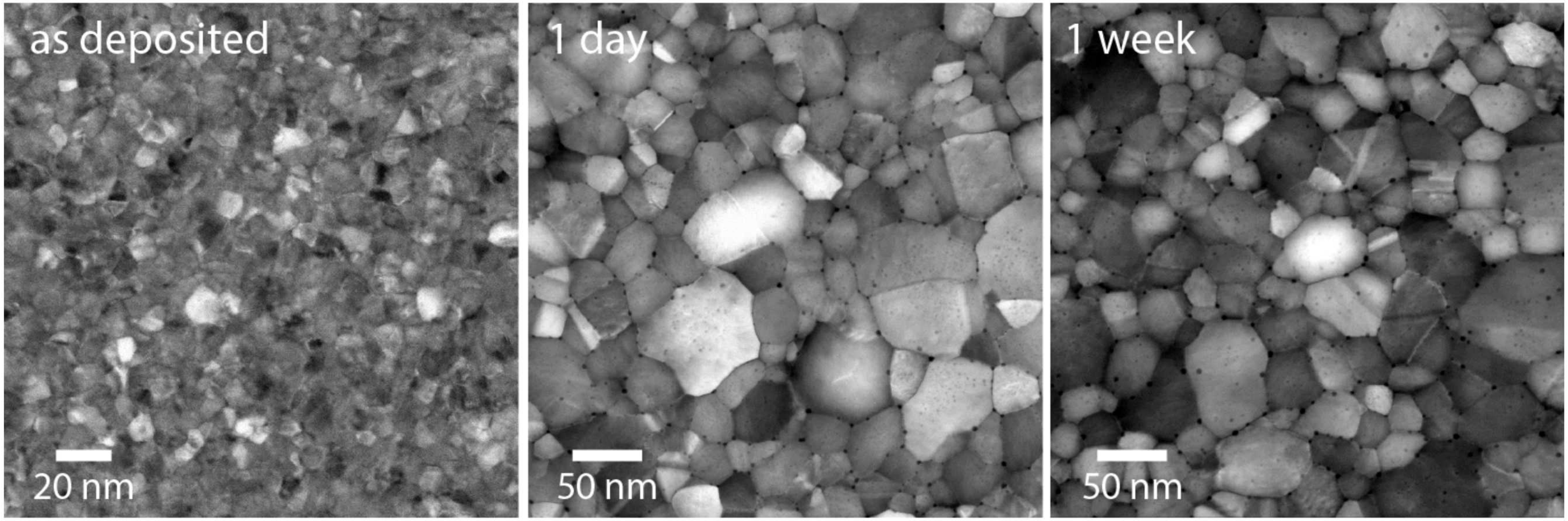
Murdoch & Schuh, ***J. Mater. Res.*** 2013.

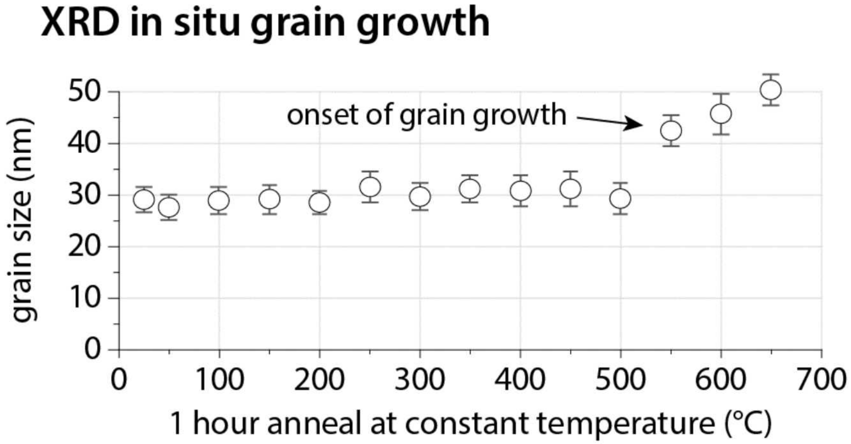
Columnar grains, and Au segregates to Pt GBs

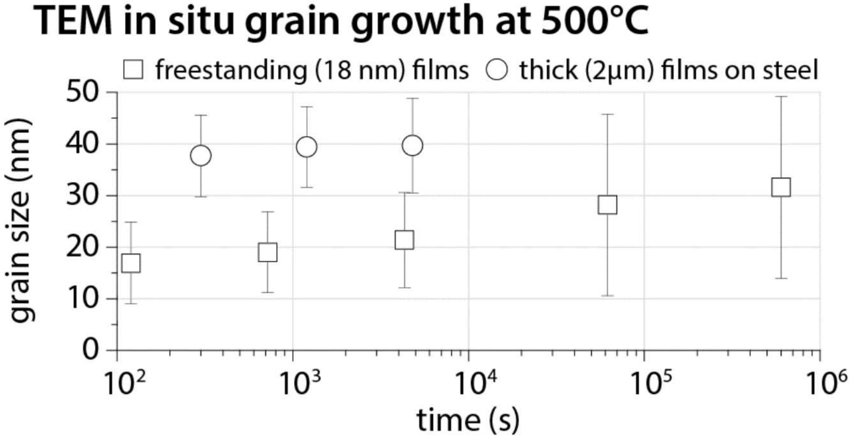


We synthesized coatings of Pt with 10% Au

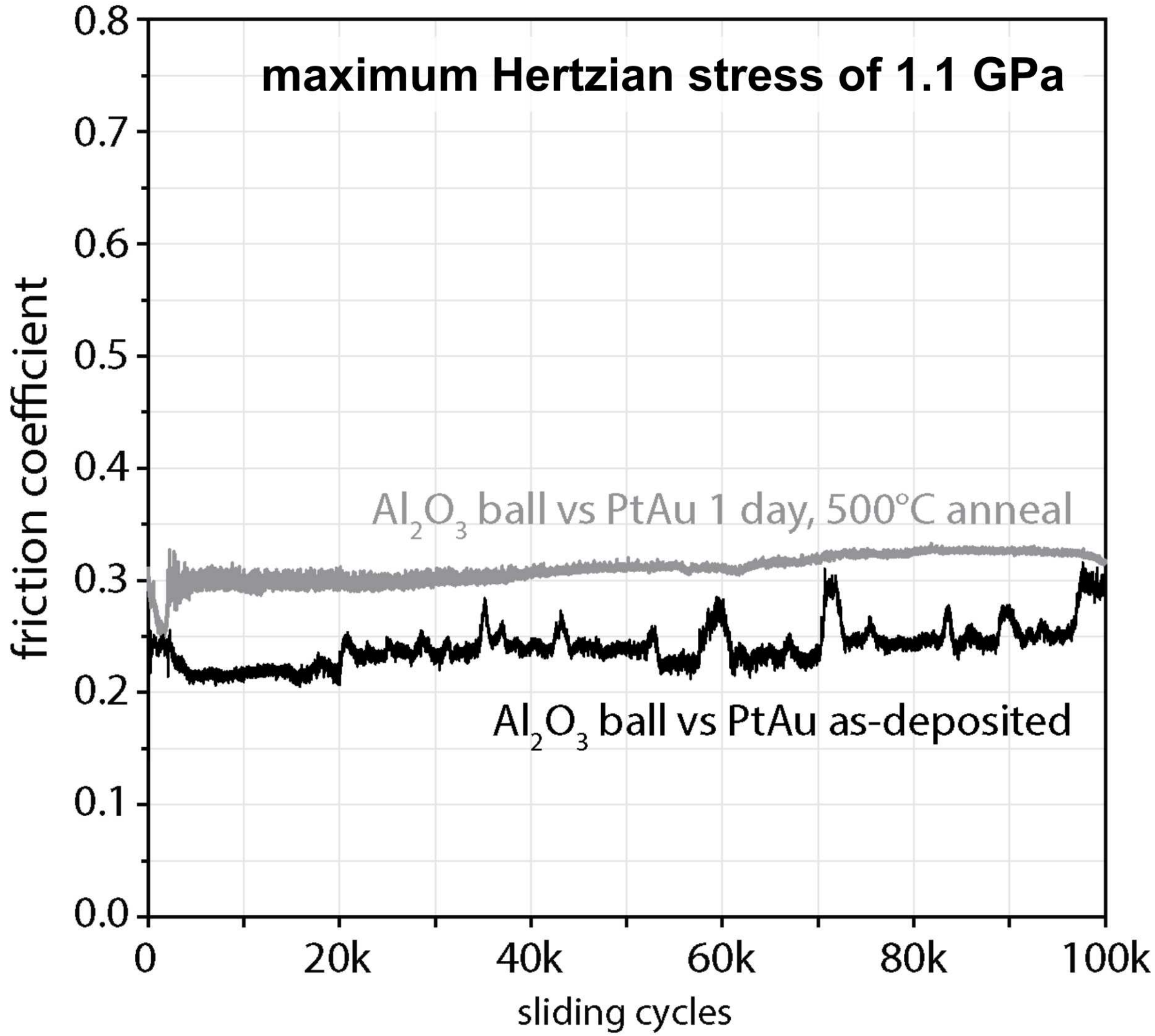
Excellent thermal stability. What about mechanical?







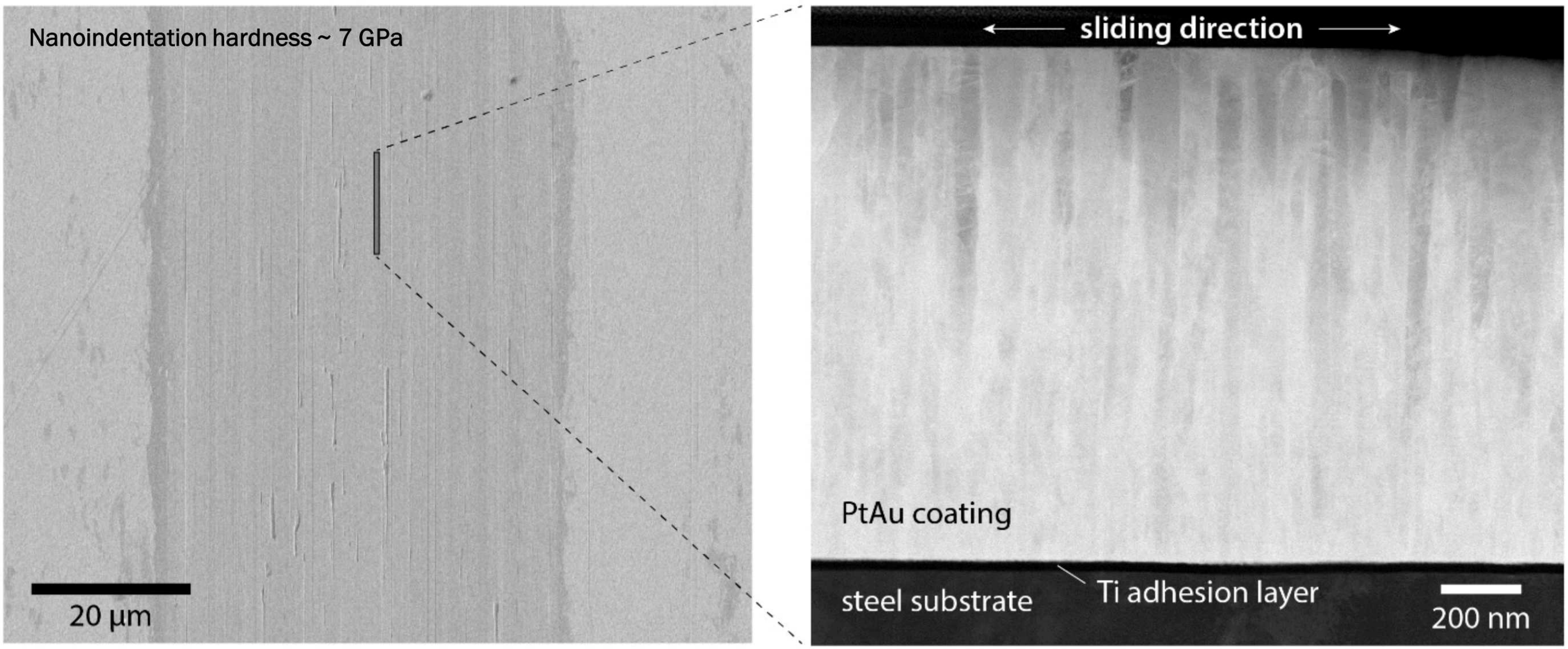
Stable nanocrystalline PtAu exhibited long-lived low friction (sliding against sapphire)



J. F. Curry, et al., ***Advanced Materials (2018)***

No evidence of microstructural evolution after prolonged tests

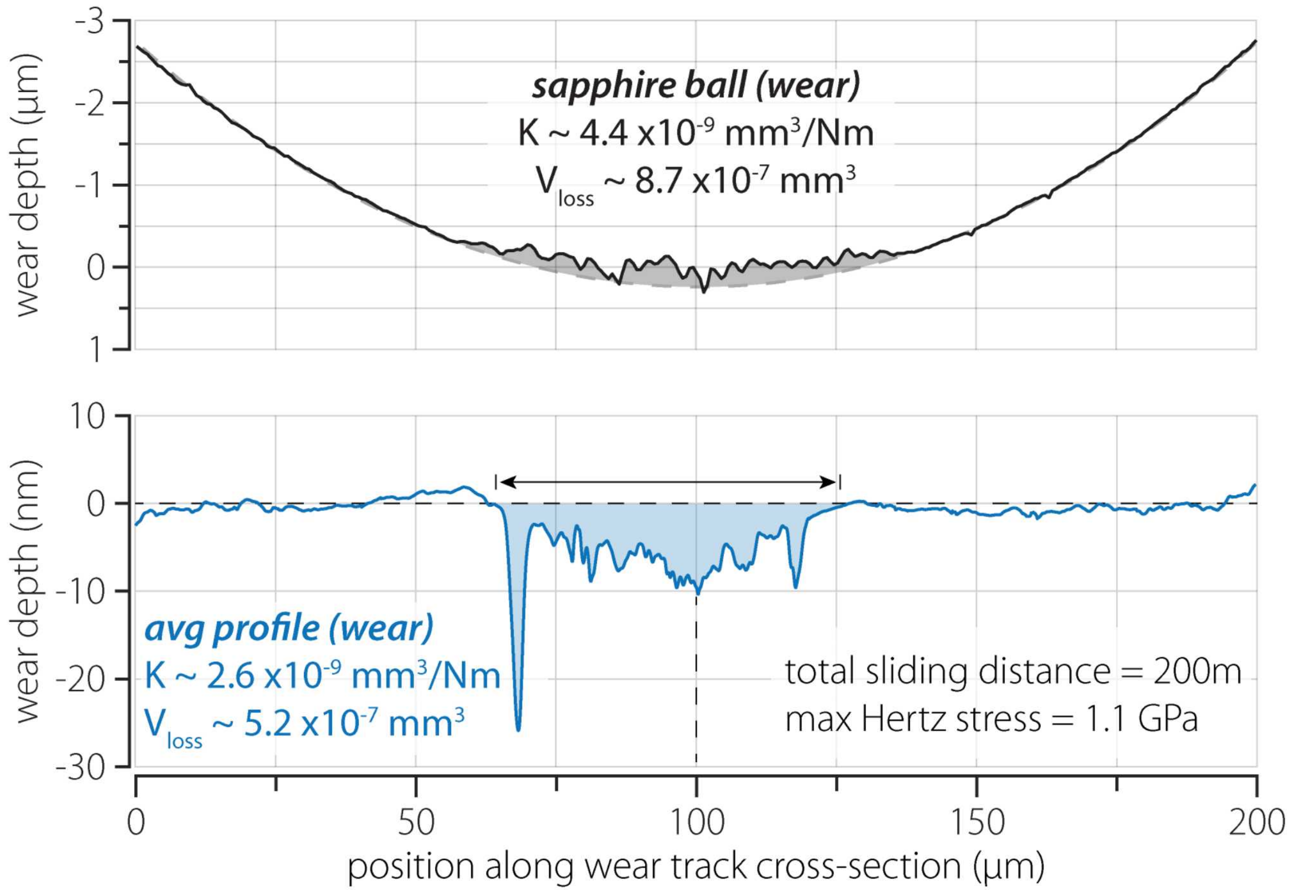
SEM of PtAu wear track after 100k passes **TEM cross-sections of wear track**

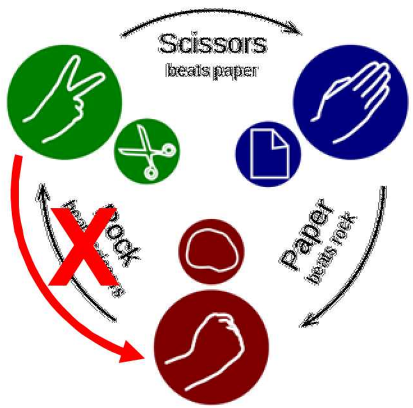


No evidence of microstructural evolution
(maximum Hertzian stress of 1.1 GPa)

J. F. Curry, et al., Advanced Materials (2018)

Wear in Cross-Section: Ultra-low wear rate of PtAu ($3x10^{-9}$ mm^3/N-m)

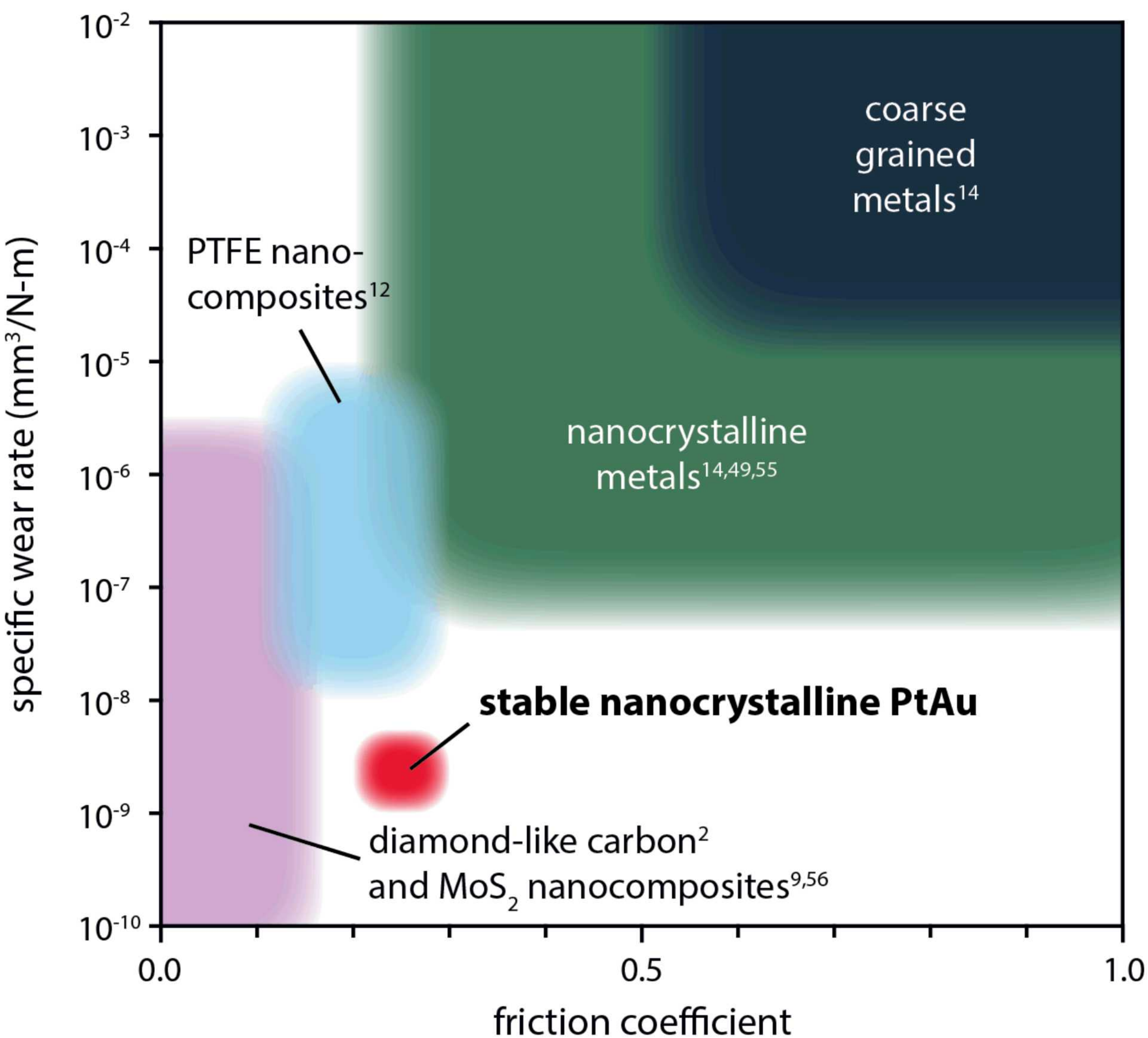




… when scissors beat rock!

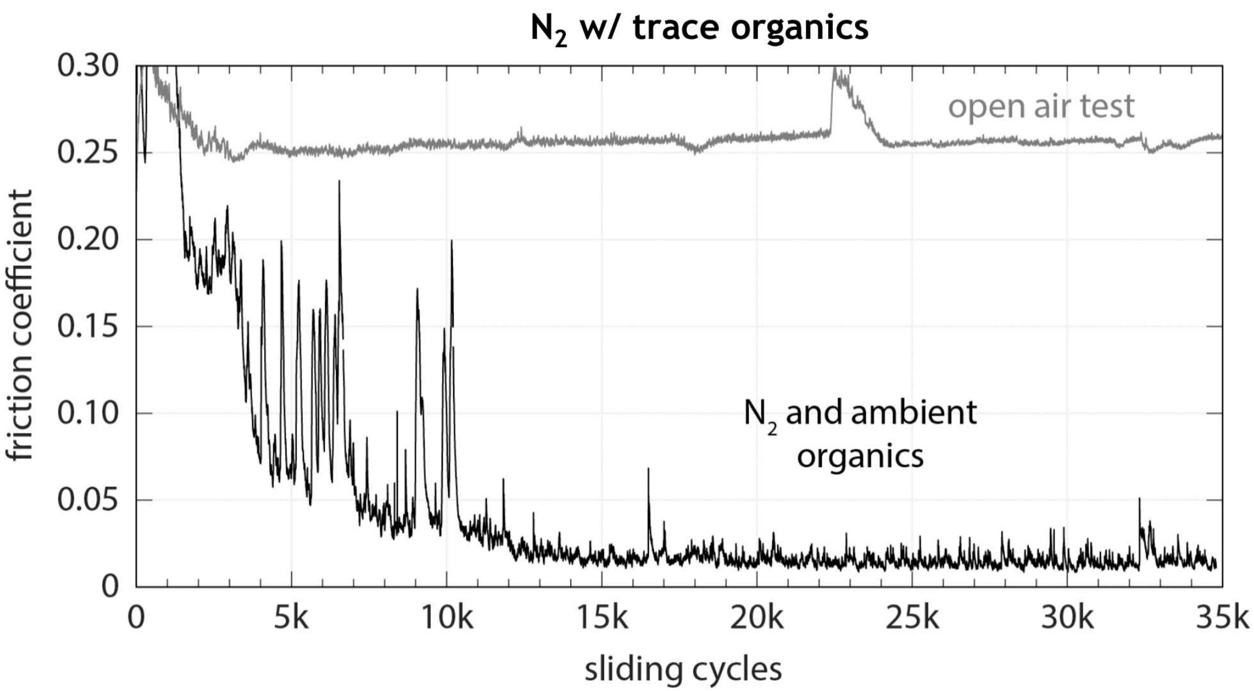
J. F. Curry, et al., ***Advanced Materials (2018)***

Significant improvement, more work to do…



J. F. Curry, et al., ***Advanced Materials (2018)***

Friction Behavior in Anaerobic Environments



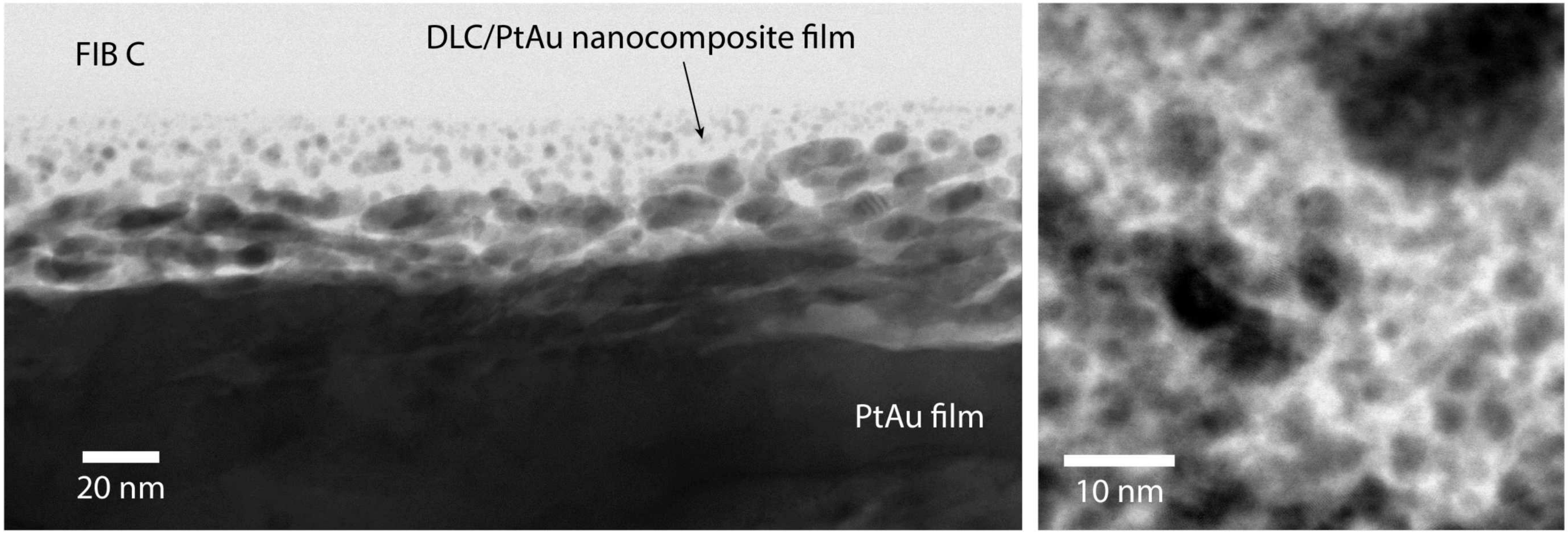
Compounds Identified
trichloroethylene
acetic acid
benzaldehyde
tridecane
phenol
acetophenone
2-heptyl-1,3-dioxolane
butylated hydroxytoluene
benzoylformic acid
phenylmaleic anhydride

(20-100 ng/L)

… in situ tribo-chemical formation of DLC films from "thin air"!
(Albuquerque is at 6,000ft/1,800m of elevation, after all…)

N. Argibay et al., Carbon (2018)

TEM Reveals a Nanocomposite Structure (DLC/PtAu Nanoparticles)



confirmed **diamond-like carbon (DLC)** using Raman analysis with 20% hydrogenation using elastic recoil

N. Argibay et al., ***Carbon (2018)***

Acknowledging the Many Contributors to this Story

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UNM/SNL

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Sandia National Labs

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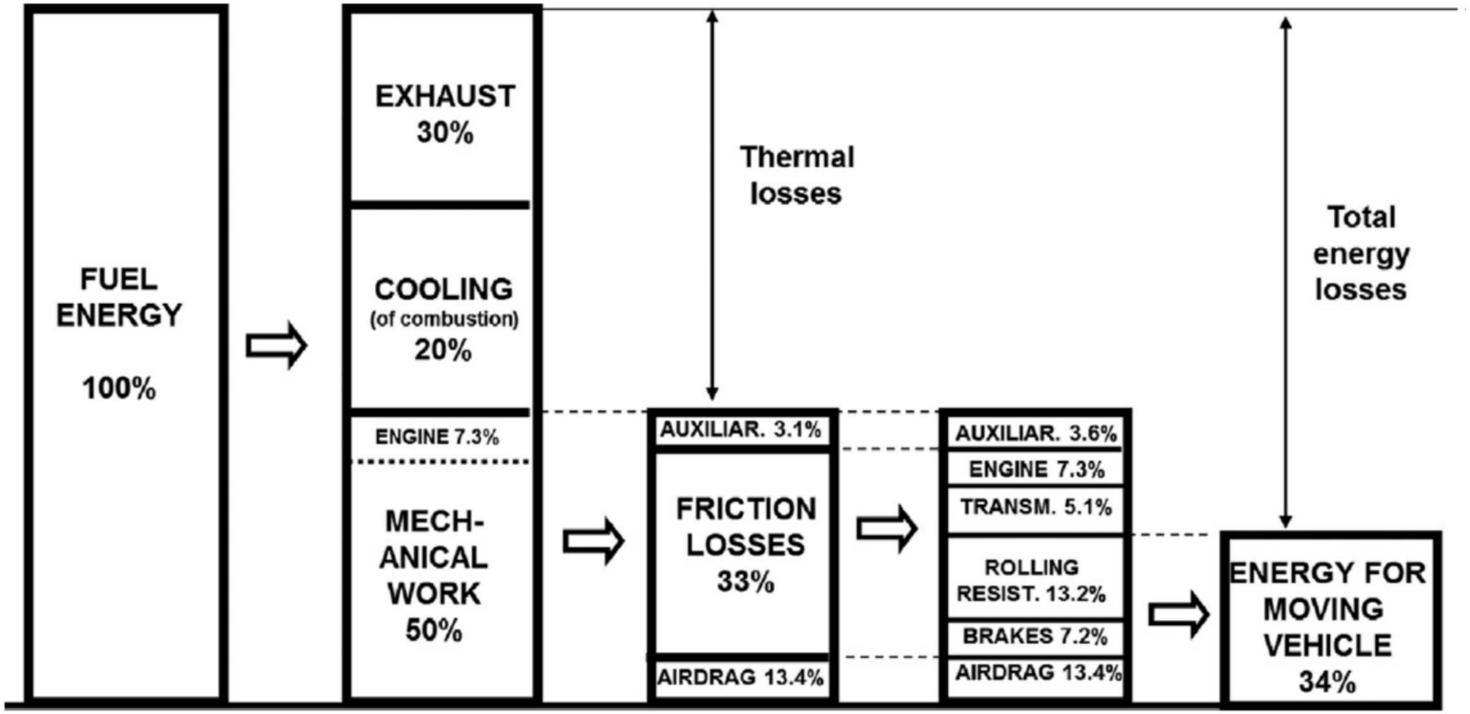
Dr. **Arthur Karyia**
Senior Staff Member
Sandia National Labs

Mr. **Justin Vanness**
Senior Staff Member
Sandia National Labs

Special thanks to **Dr. Michael Chandross**
(who is typically a co-presenter…)

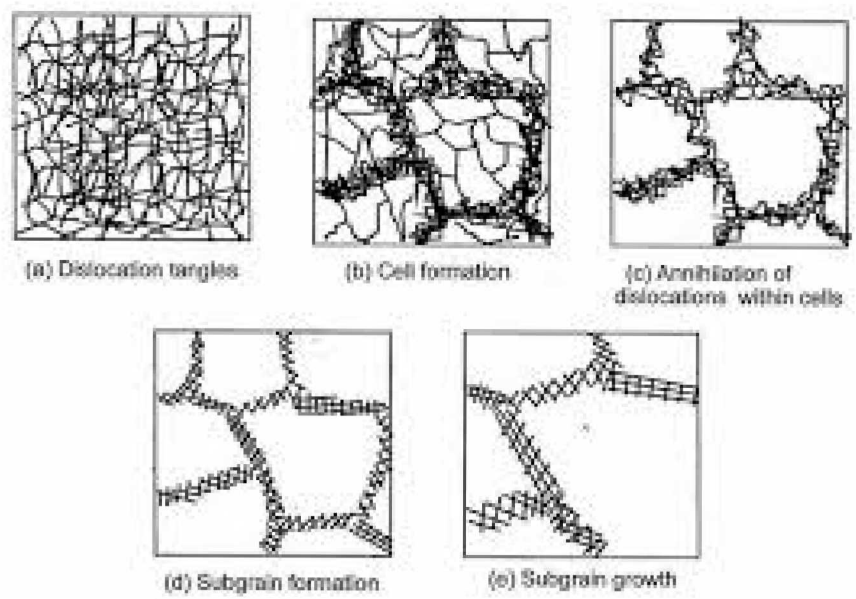
Questions?

Transportation – Passenger Car Energy Losses



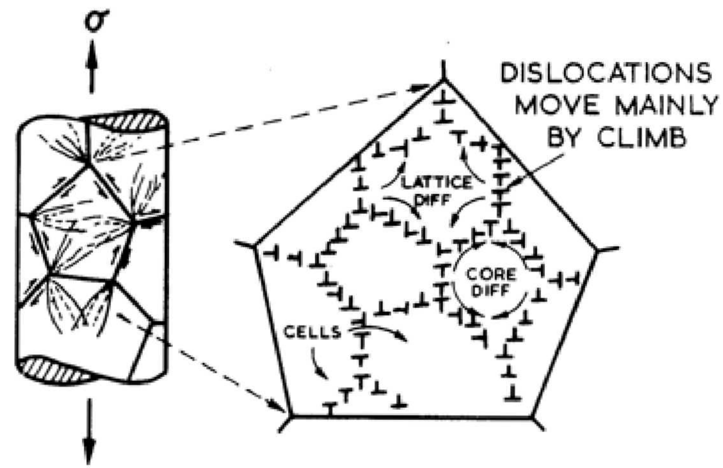
ref: K. Holmberg et al., Tribo. Int. 2012

Competition between refinement and coarsening

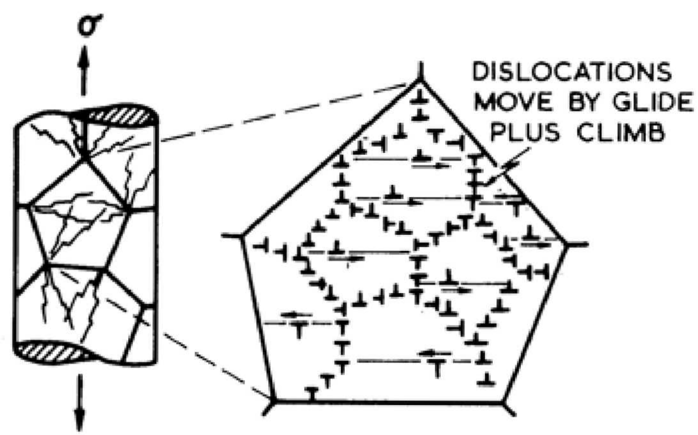


http://engineering.dartmouth.edu/defmech/chapter_2.htm

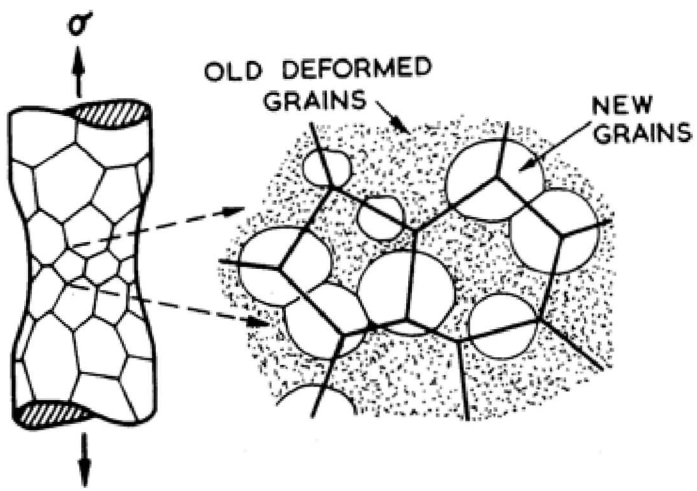
creep:



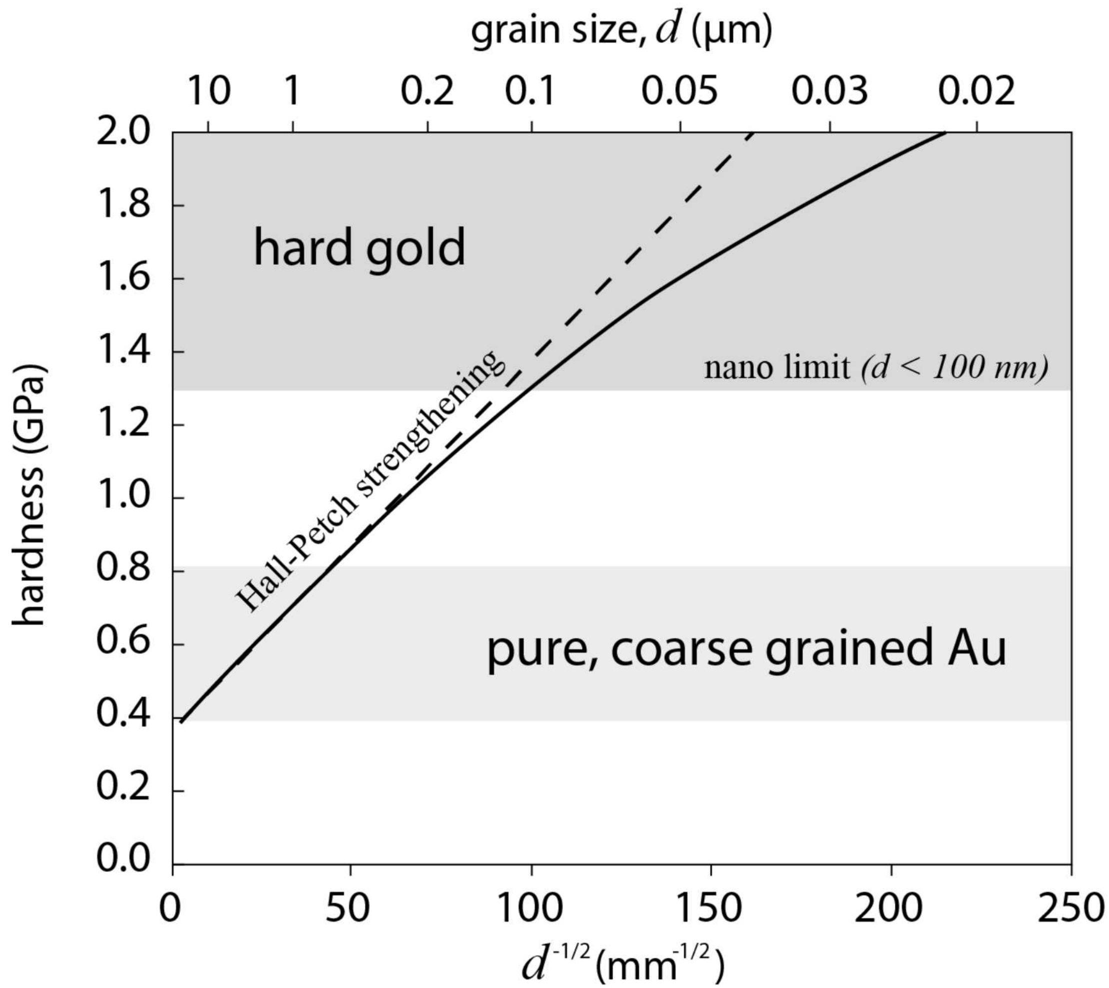
glide:



DRX:



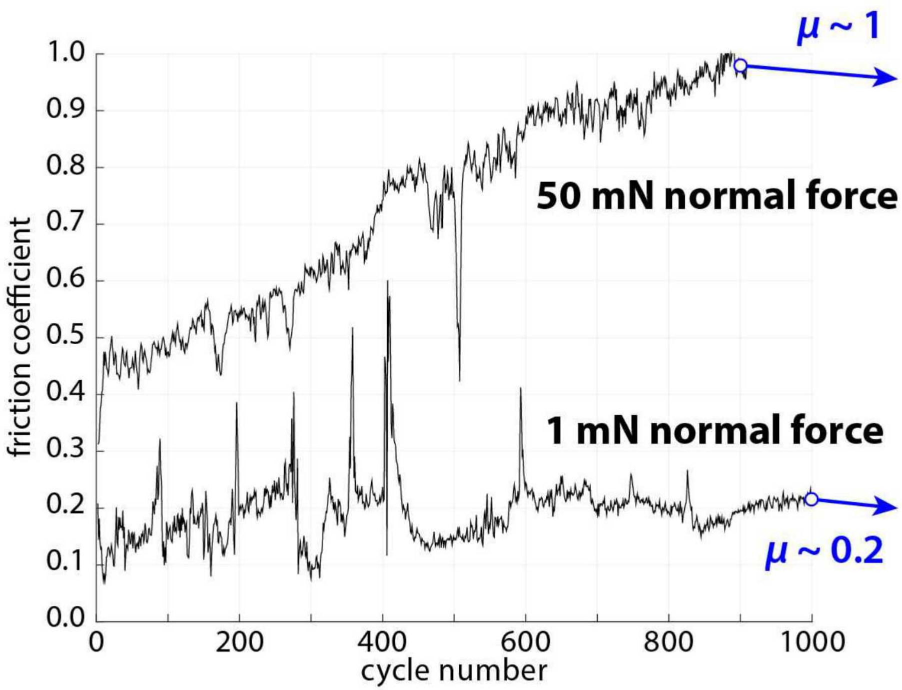
Grain-size dependent strength: Hall-Petch



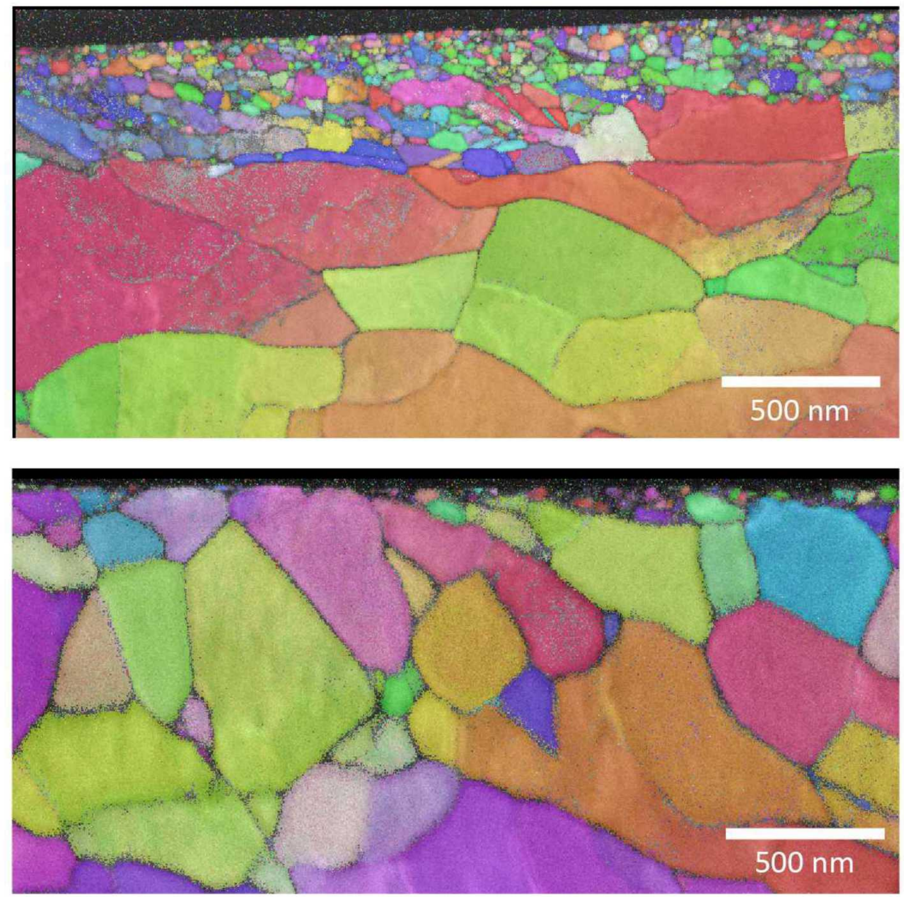
Reference: C. Lo, J. Augis, and M. Pinnel, JAP (1979)

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)

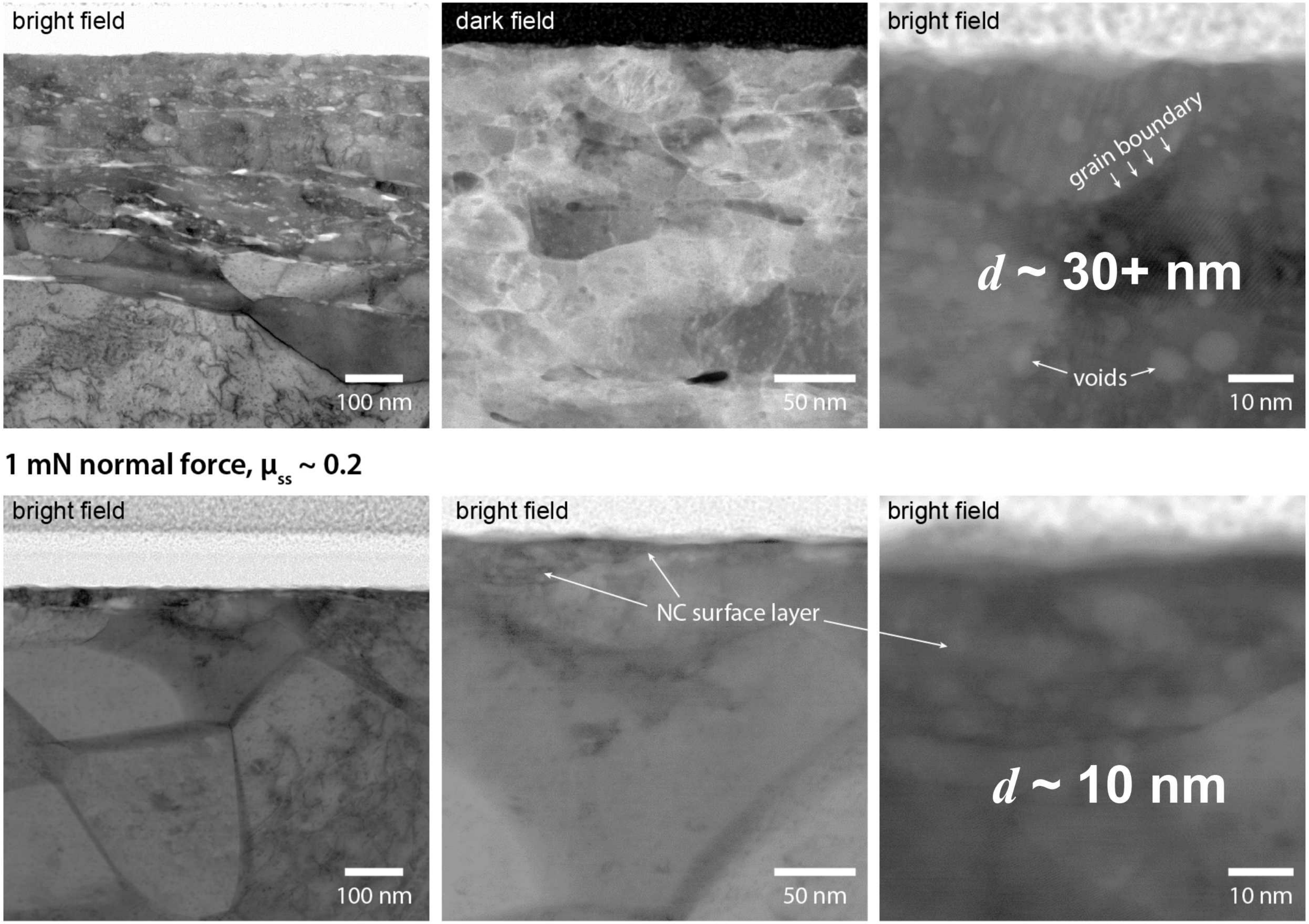


Again we see fine grain size in both cases…
but the low friction case seems smaller.

Microstructurally small differences in grain size = BIG difference in friction response

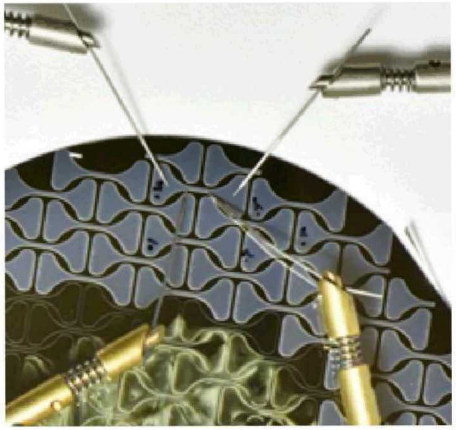
50 mN normal force, μ_{final} ~ 1.0

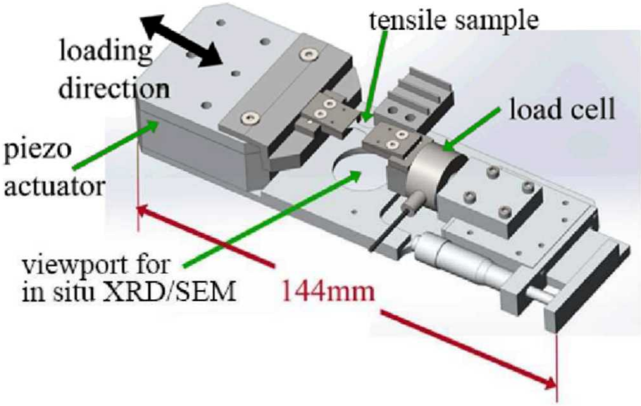
1 mN normal force, μ_{ss} ~ 0.2

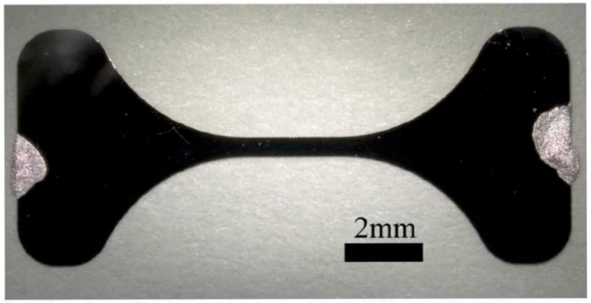


Tensile and fatigue properties of PtAu – Experiments & Simulations

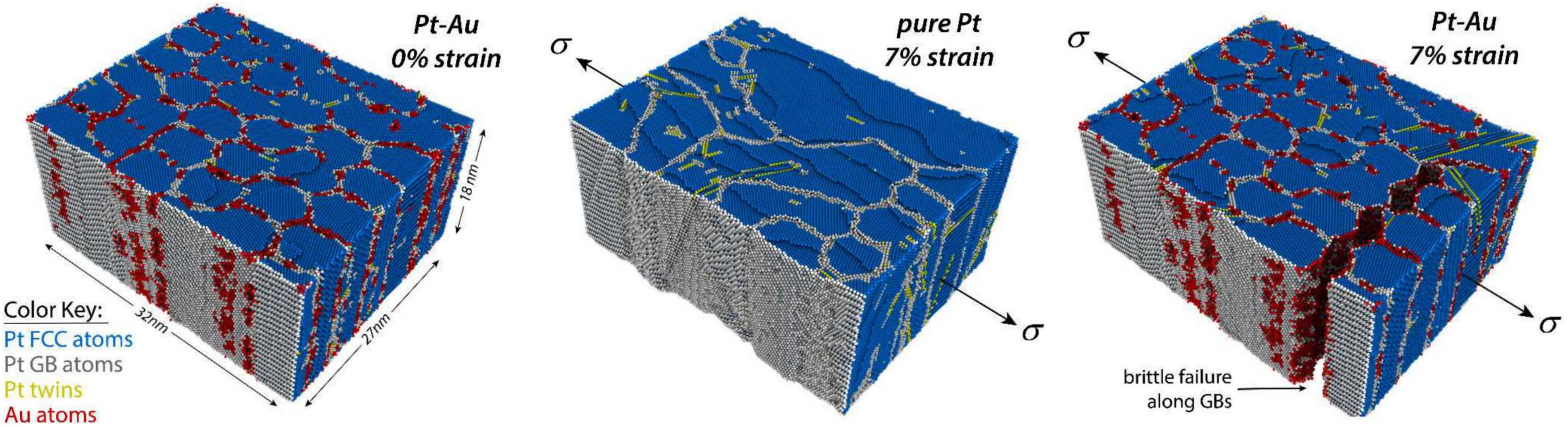
Tensile Testing (Fatigue and Yield Strength):





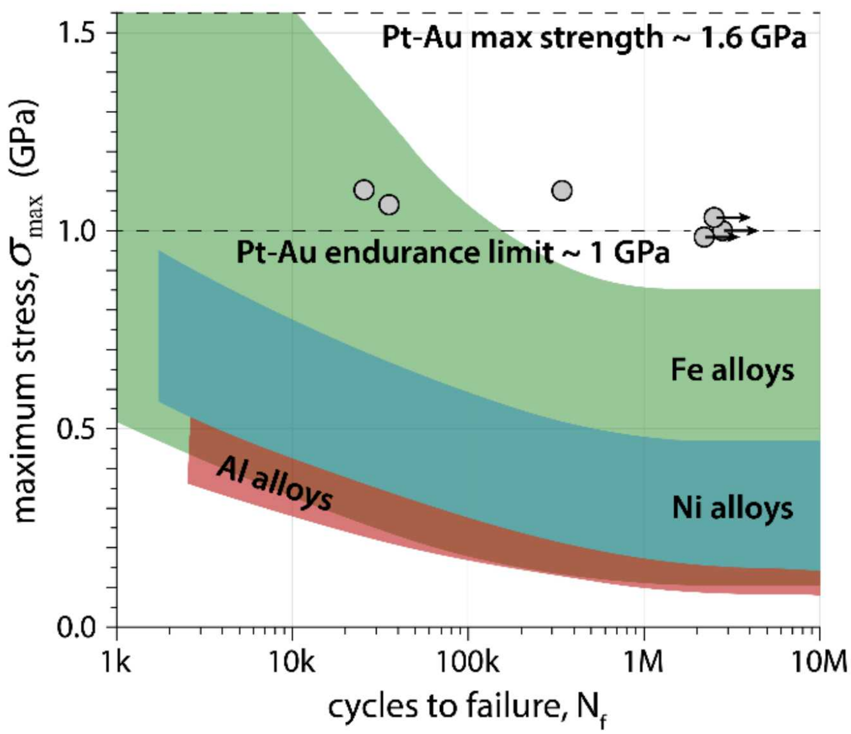


MD Simulation Snapshots (grain growth during uniaxial tensile stress exposure):

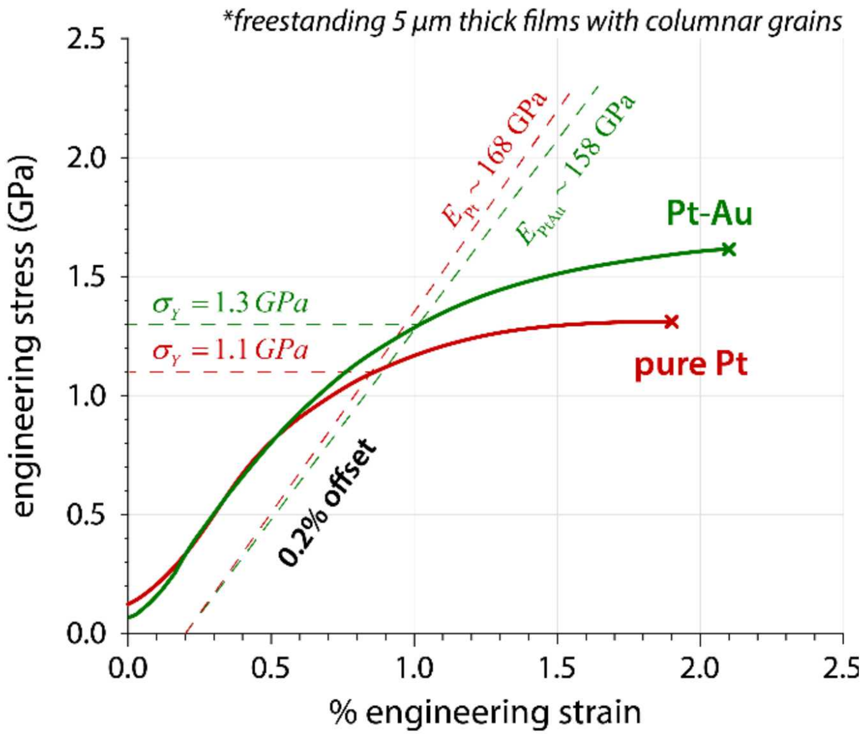


PtAu alloys have high yield strength and remarkable fatigue resistance

Experimental Tensile Fatigue:



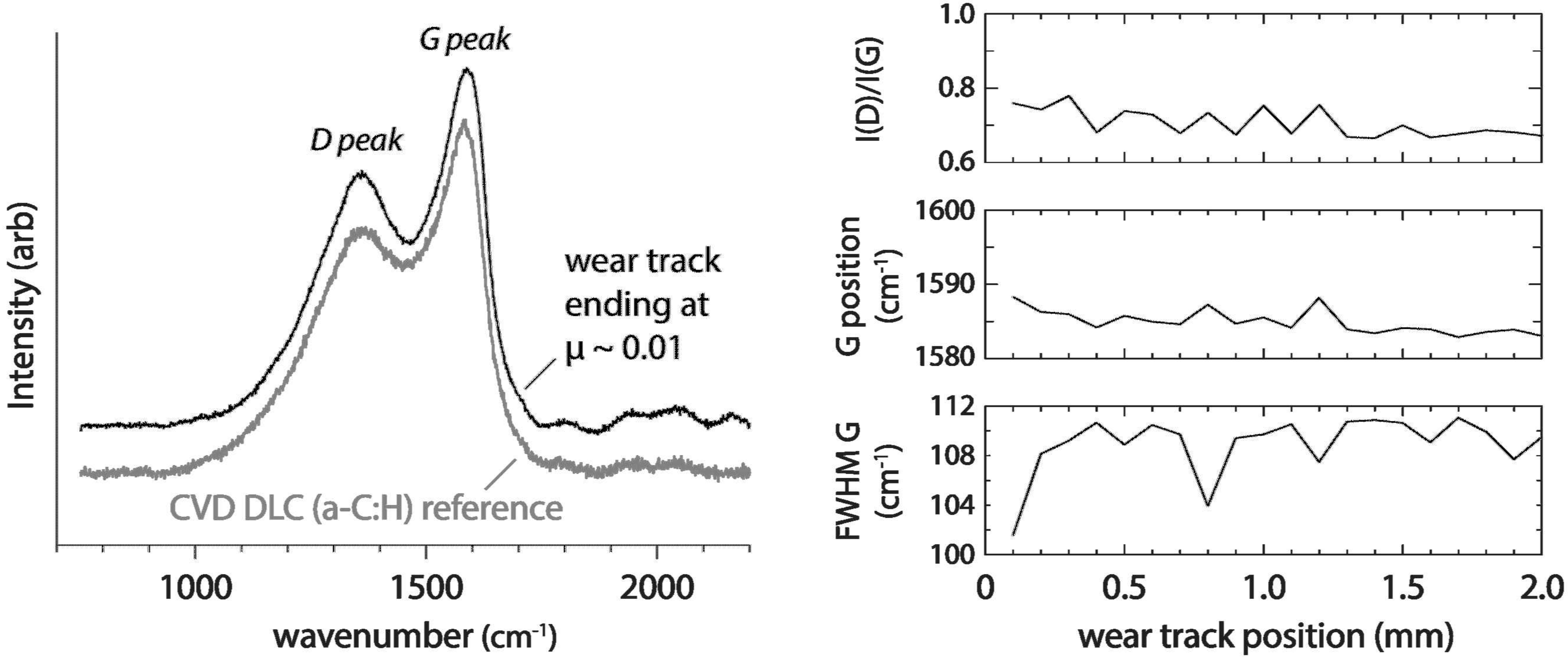
Experimental stress-strain data:



This high yield strength and fatigue resistance generated an interesting result…

J. F. Curry, et al., ***Advanced Materials (2018)***

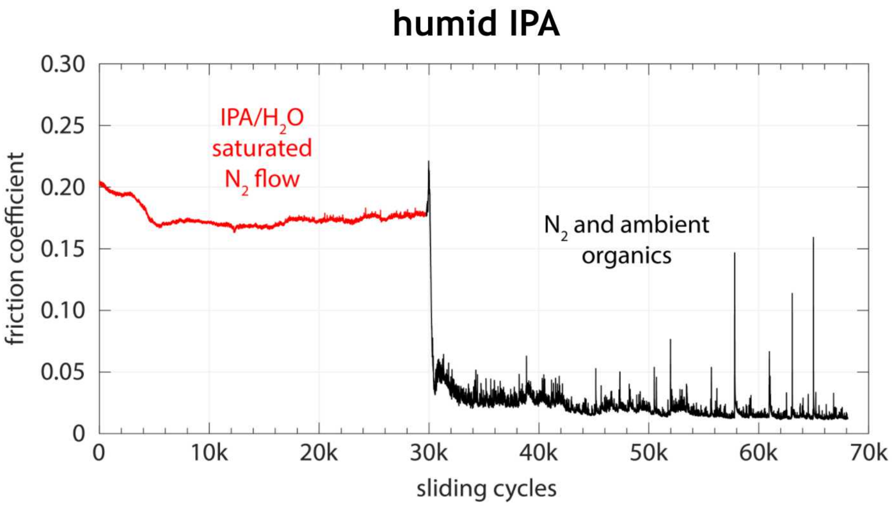
Raman Spectroscopy -- Highly Graphitic Hydrogenated (20%) DLC

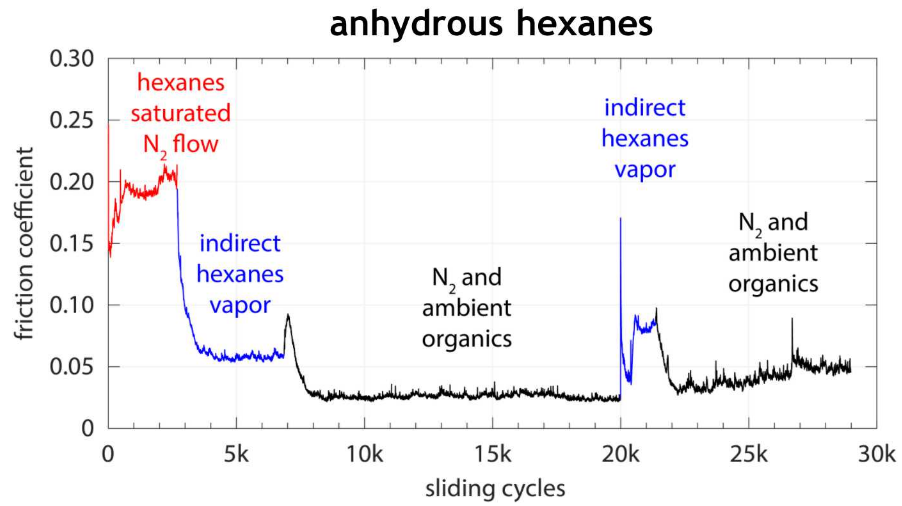


elastic recoil showed ~ 20% H

N. Argibay et al., ***Carbon (2018)***

Friction behavior directly linked to hydrocarbon concentration (dry)

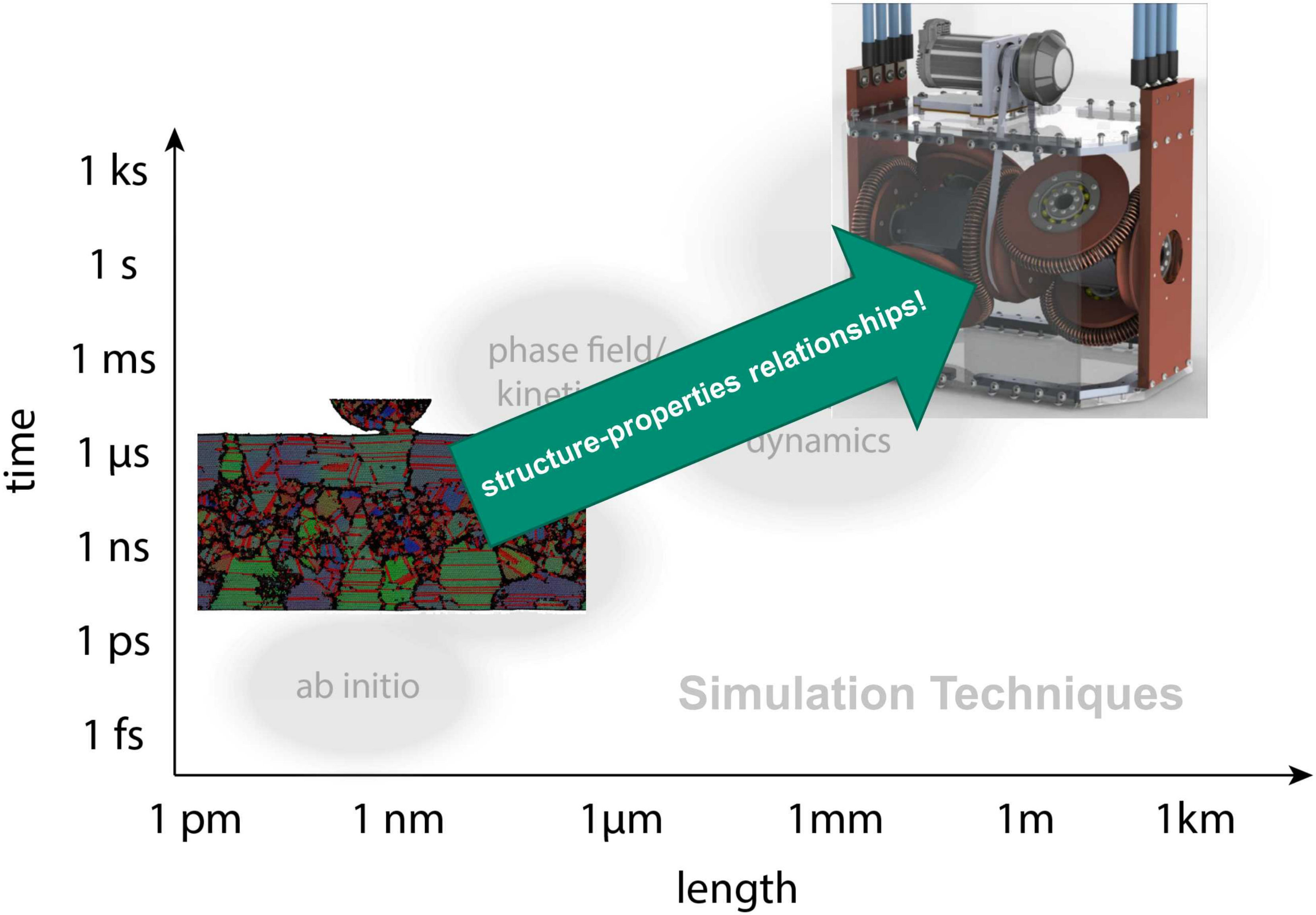




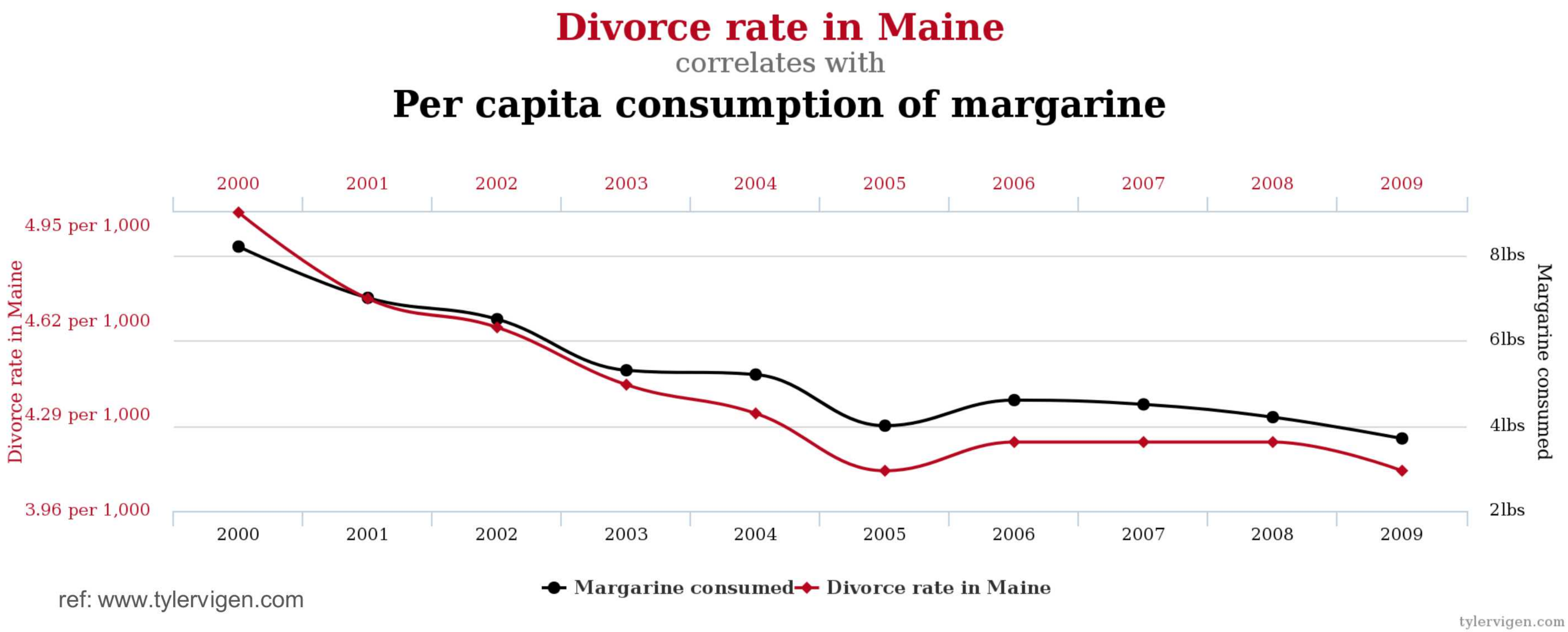
Alcohol helps, but only in moderation.

*N. Argibay et al., **Carbon (2018)***

Bridging the Gap From Atomistic Modeling to Macroscale Applications



… as we know, correlation is not causation…

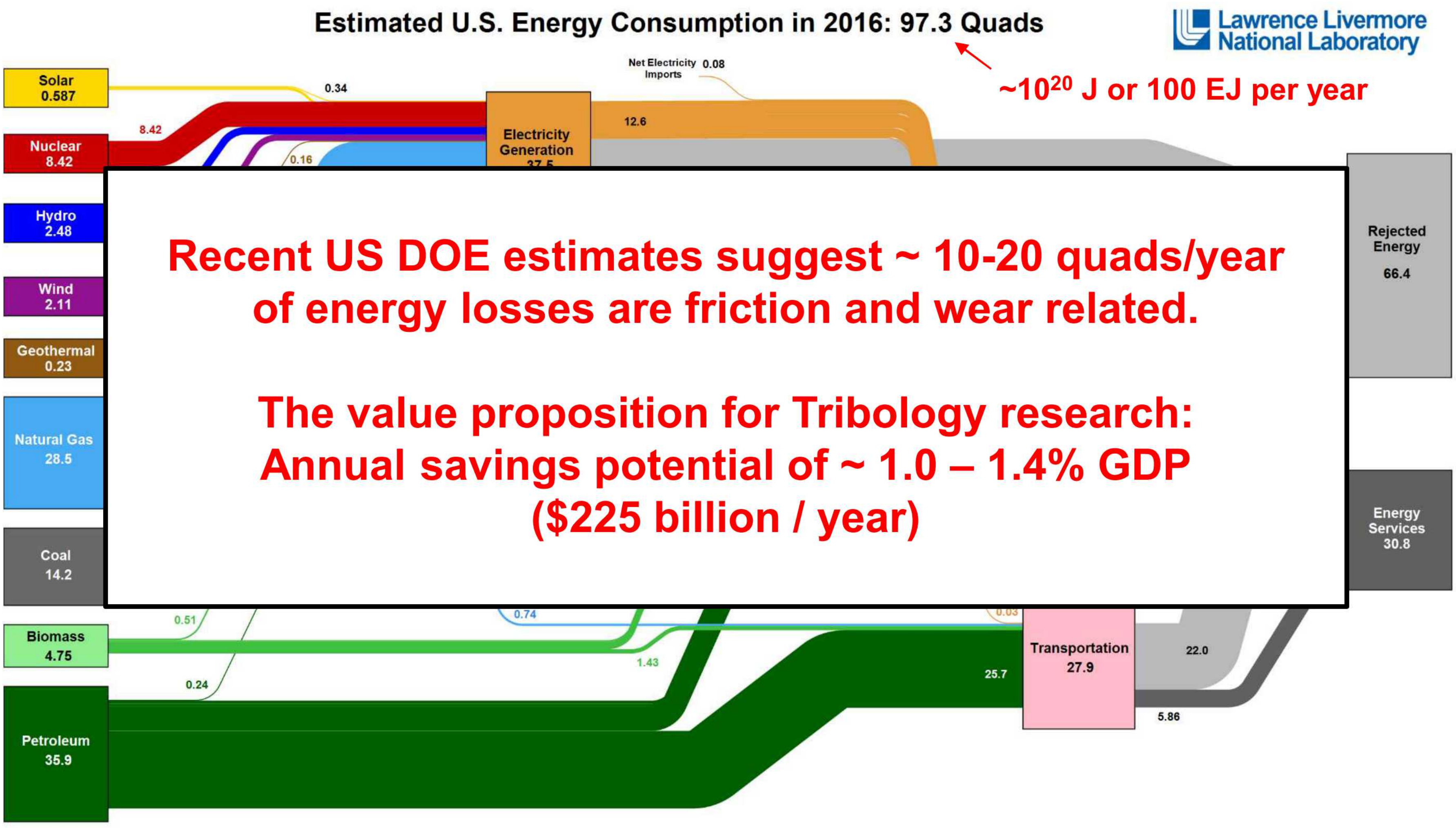


ref: www.tylervigen.com

so please, for the sake of married people in Maine…

… use only the pure stuff

Some Perspective on the Value Proposition of Tribology Research

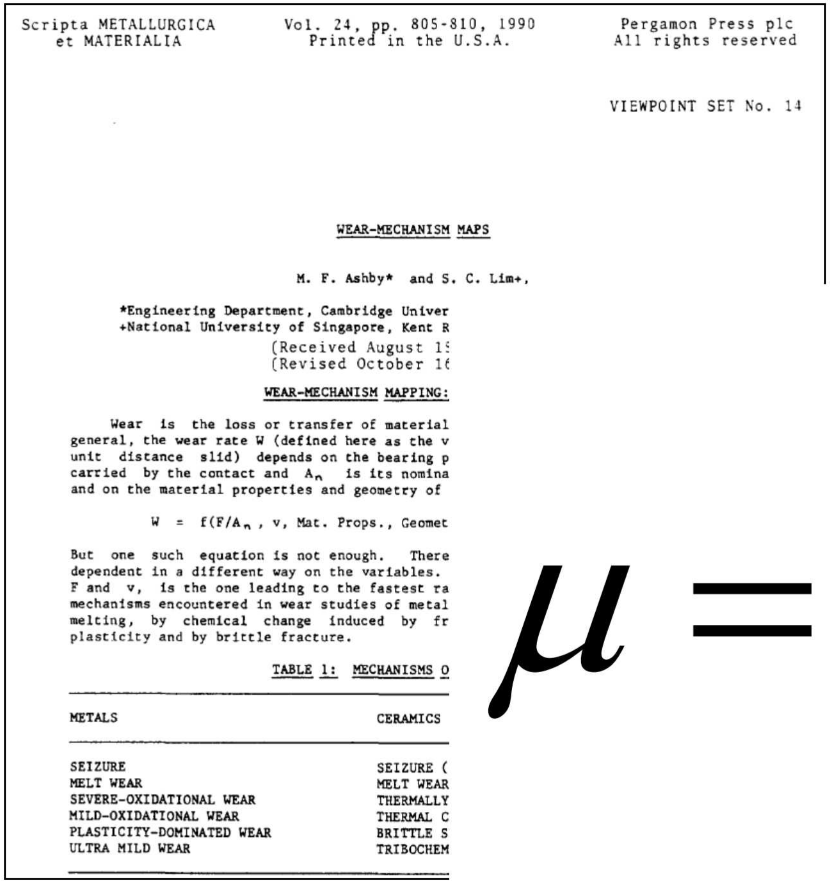


Recent US DOE estimates suggest ~ 10-20 quads/year of energy losses are friction and wear related.

The value proposition for Tribology research: Annual savings potential of ~ 1.0 – 1.4% GDP ($225 billion / year)

Source: LLNL March, 2017. Data is based on DOE/EIA MER (2016). If this information or a reproduction of it is used, credit must be given to the Lawrence Livermore National Laboratory and the Department of Energy, under whose auspices the work was performed. This chart was revised in 2017 to reflect changes made in mid-2016 to the Energy Information Administration's analysis methodology and reporting. The efficiency of electricity production is calculated as the total retail electricity delivered divided by the primary energy input into electricity generation. End use efficiency is estimated as 65% for the residential sector, 65% for the commercial sector, 21% for the transportation sector, and 49% for the industrial sector which was updated in 2017 to reflect DOE's analysis of manufacturing. Totals may not equal sum of components due to independent rounding. LLNL-MI-410527

Attempts to define wear & friction regimes remain empirical/phenomenological



Scripta METALLURGICA et MATERIALIA
Vol. 24, pp. 805-810, 1990
Printed in the U.S.A.
Pergamon Press plc

VIEWPOINT SET No. 14

WEAR-MECHANISM MAPS

M. F. Ashby* and S. C. Lim+,

*Engineering Department, Cambridge Univer
+National University of Singapore, Kent R
(Received August 1
(Revised October 16

WEAR-MECHANISM MAPPING:

Wear is the loss or transfer of material
general, the wear rate W (defined here as the v
unit distance slid) depends on the bearing p
carried by the contact and A_n is its nomina
and on the material properties and geometry of

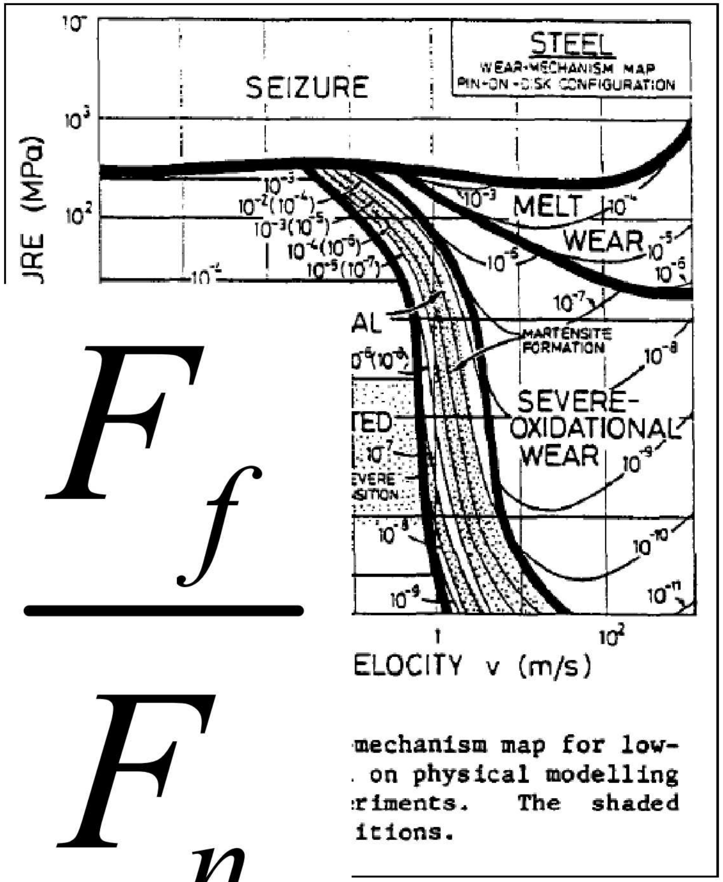
W = f(F/A_n , v, Mat. Props., Geomet

But one such equation is not enough. There
dependent in a different way on the variables.
F and v, is the one leading to the fastest ra
mechanisms encountered in wear studies of metal
melting, by chemical change induced by fr
plasticity and by brittle fracture.

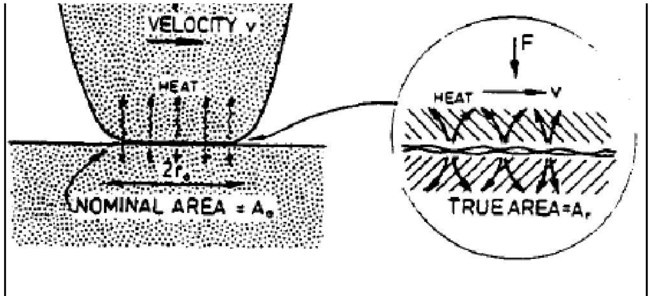
TABLE 1: MECHANISMS O

METALS	CERAMICS
SEIZURE	SEIZURE (
MELT WEAR	MELT WEAR
SEVERE-OXIDATIONAL WEAR	THERMALLY
MILD-OXIDATIONAL WEAR	THERMAL C
PLASTICITY-DOMINATED WEAR	BRITTLE S
ULTRA MILD WEAR	TRIBOCHEM

$$\mu = \frac{F_f}{F_n}$$

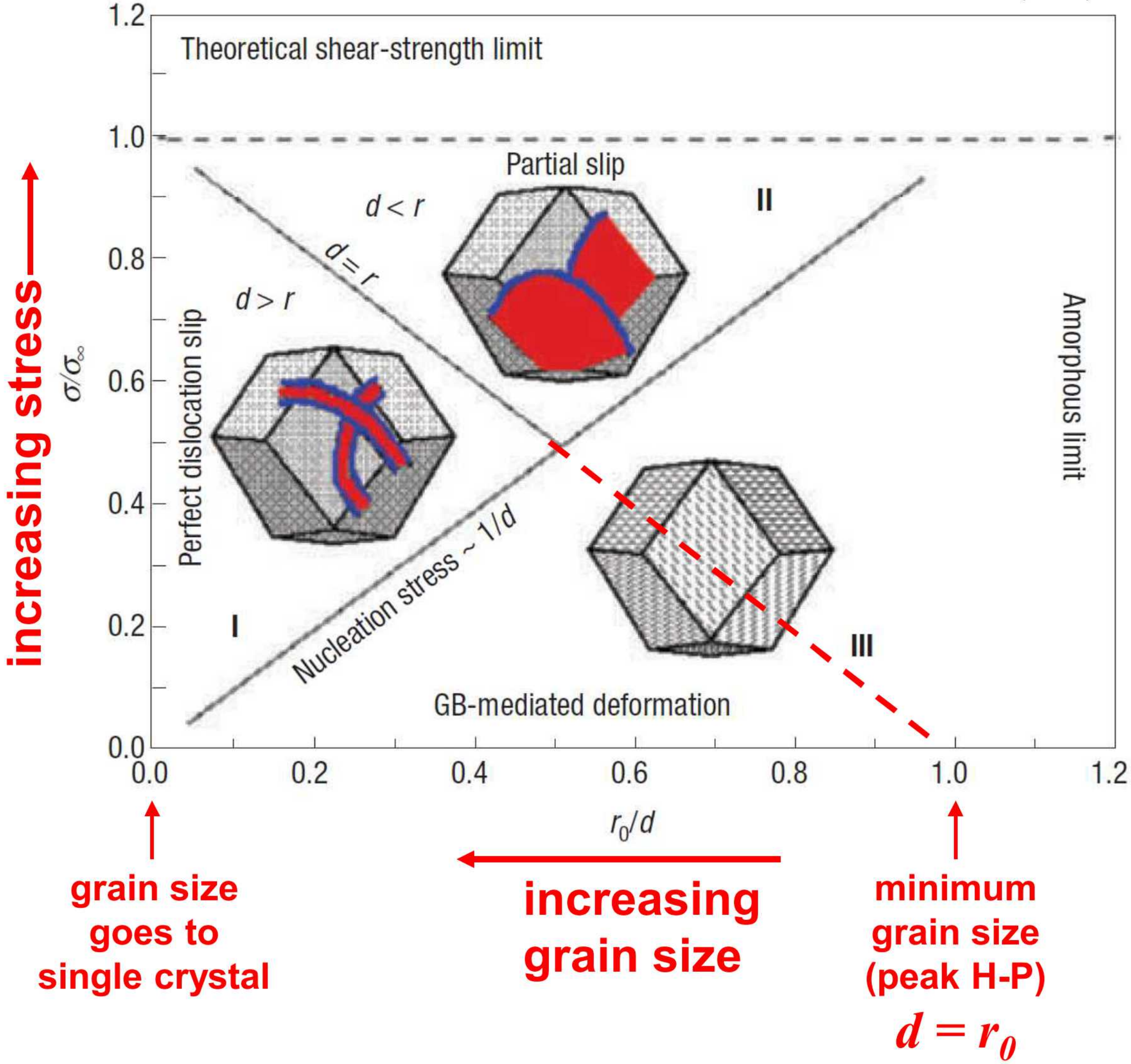


mechanism map for low-
. on physical modelling
riments. The shaded
itions.



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

ref: Yamakov et al., Nat. Mat. (2004)



Equilibrium (zero stress) dislocation splitting distance:

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

Stress-dependent splitting distance:

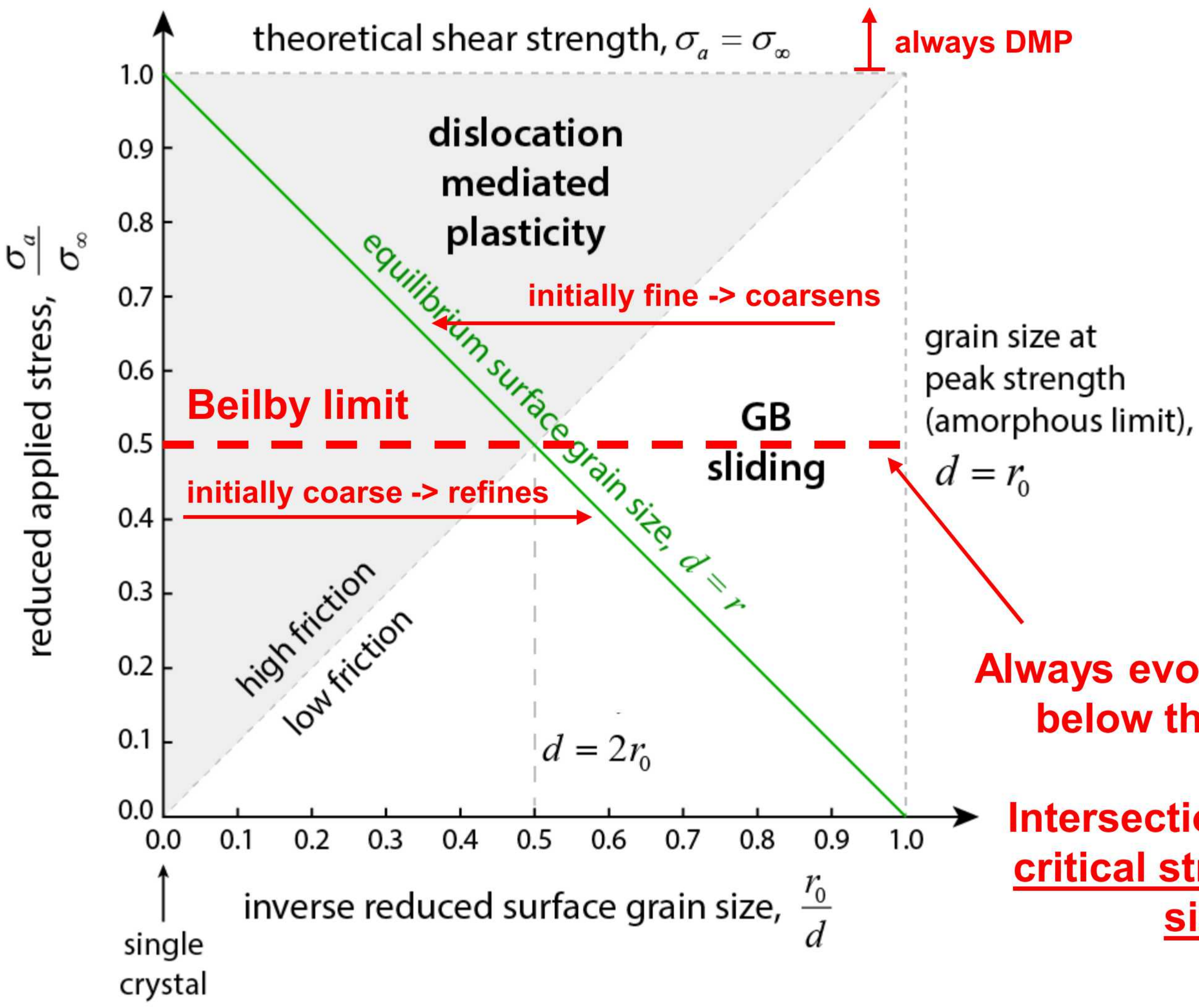
$$r = \frac{r_0}{1-\sigma_a/\sigma_\infty}$$

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 (& time-dep)

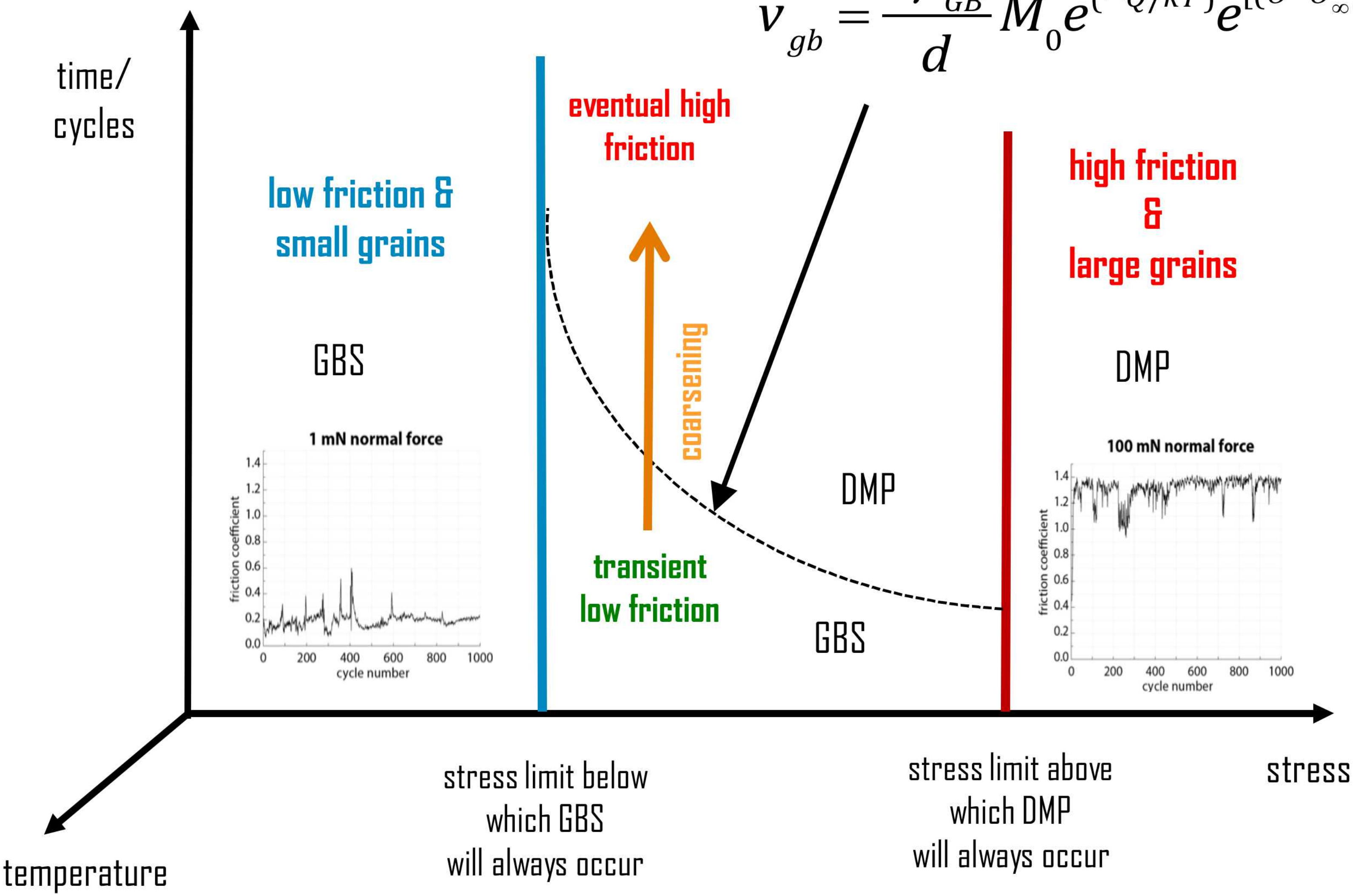


Always evolves to GBS below this stress

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

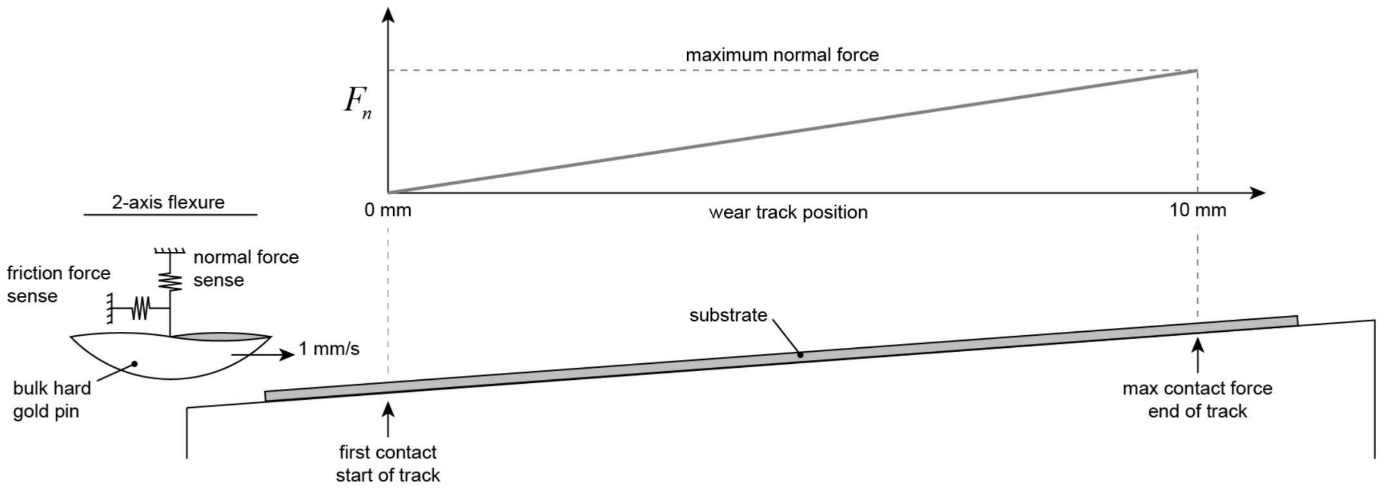
Generalized friction regimes map for metals

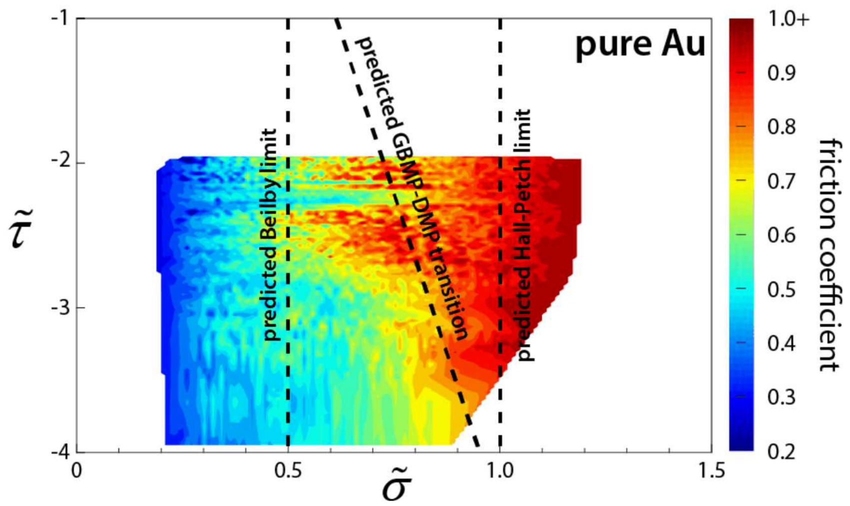
$$v_{gb} = \frac{2\gamma_{GB}}{d} M_0 e^{(-Q/kT)} e^{[(\sigma-\sigma_{\infty}/2)V^*/kT]}$$

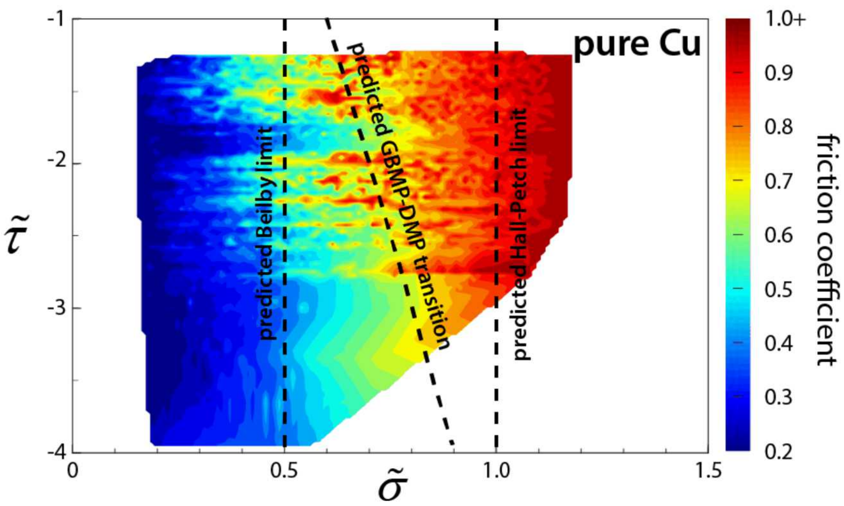


N. Argibay, et al., ***J. Materials Science (2017)***

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



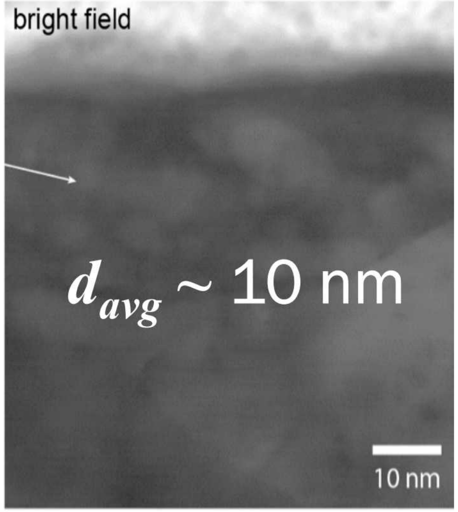


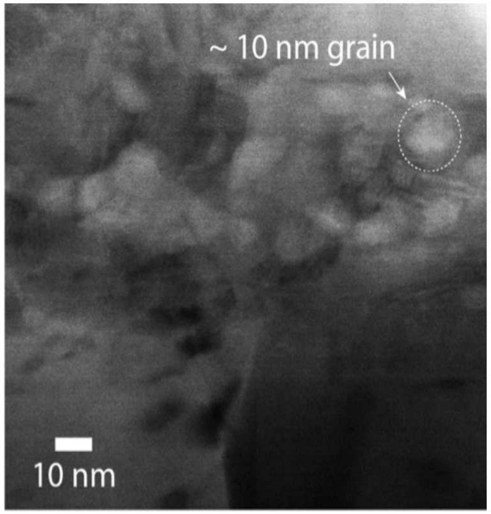


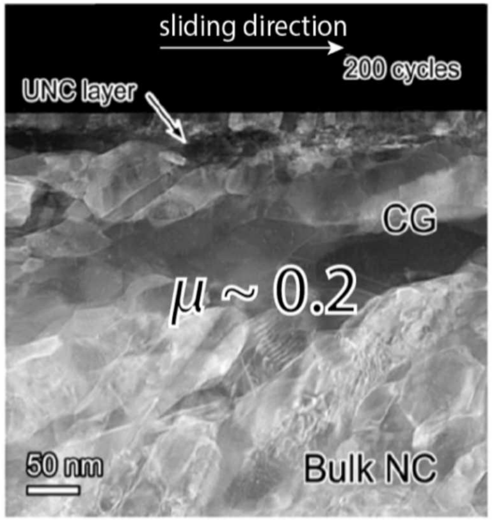
We can predict a crossover critical grain size

gold on gold	copper on copper	Si_3N_4 on nickel

low friction





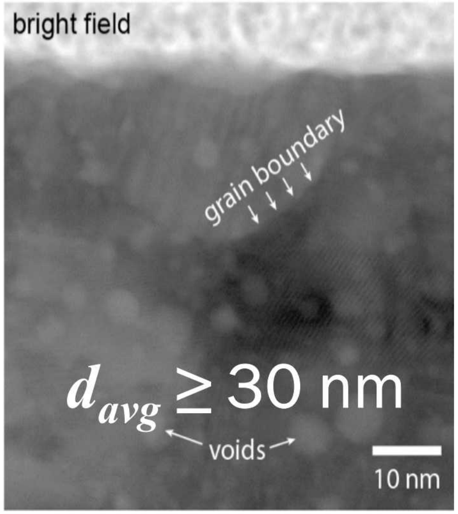


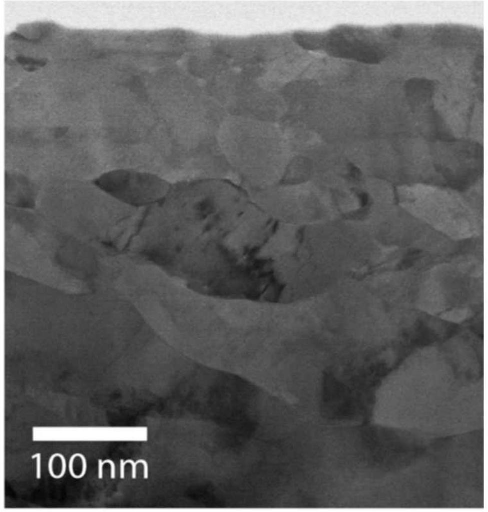
critical grain size = *17.4 nm*

critical grain size = *11.8 nm*

critical grain size = *9.8 nm*

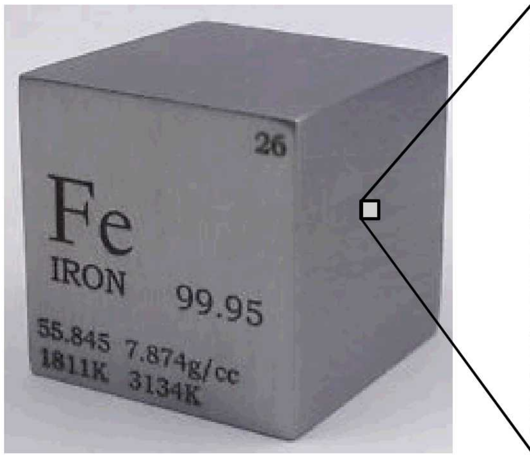
high friction



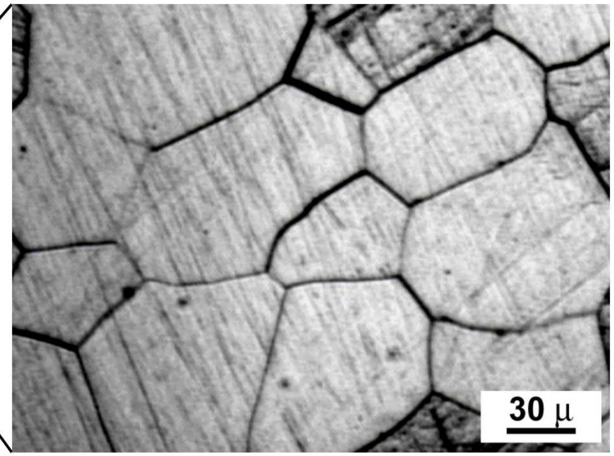


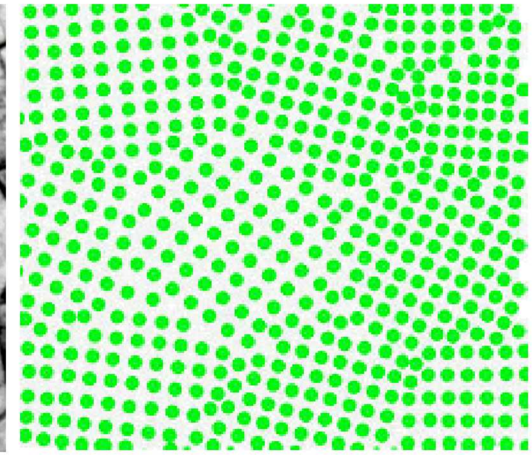
Prasad et al., *Scripta Materialia (2011)*

Crystal Structure of Metals (A Primer)



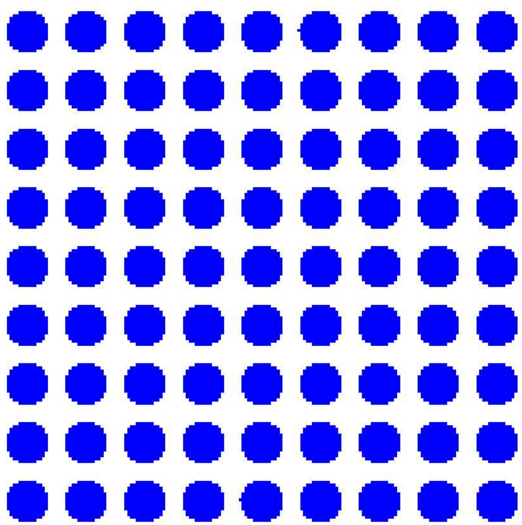
https://www.amazon.com/25-4mm-Metal-99-95-Engraved-Periodic/dp/B06WVQ7JXD





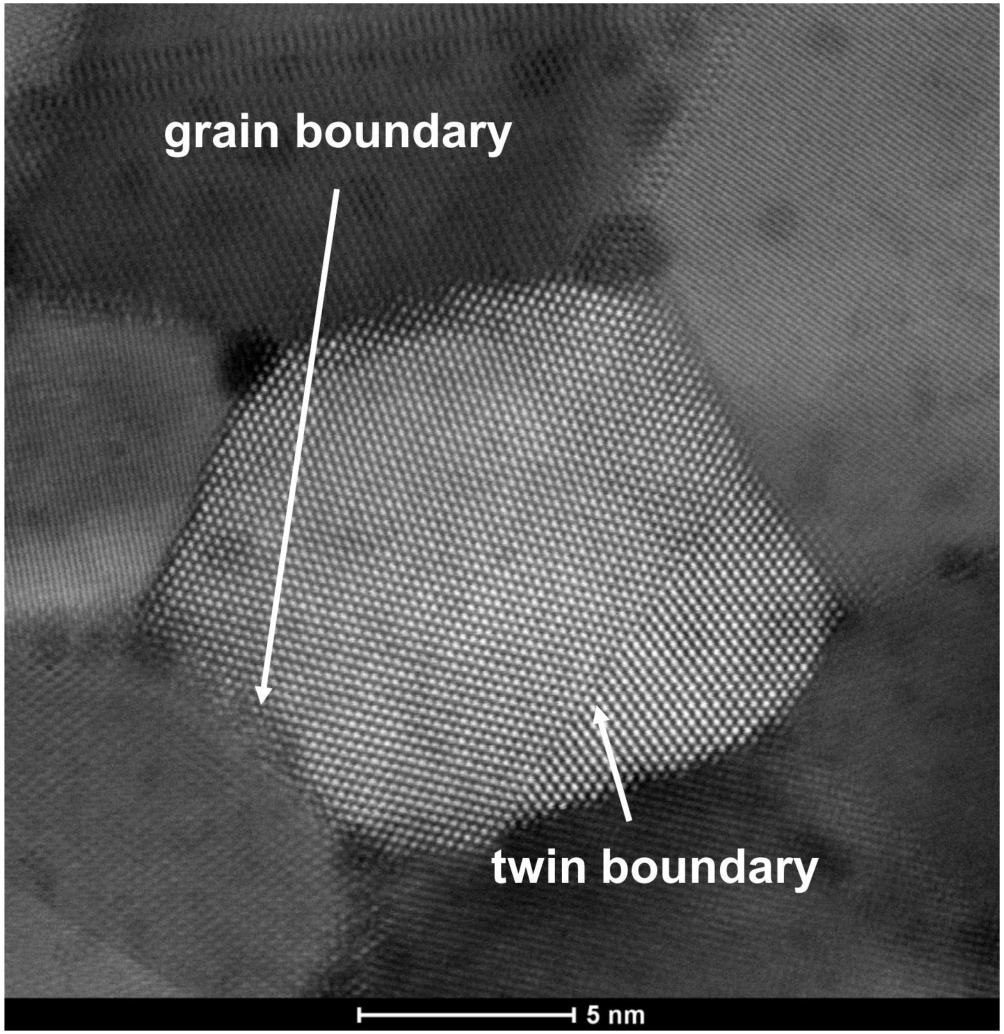
https://en.wikipedia.org/wiki/Grain_boundary

Shear via dislocation motion:

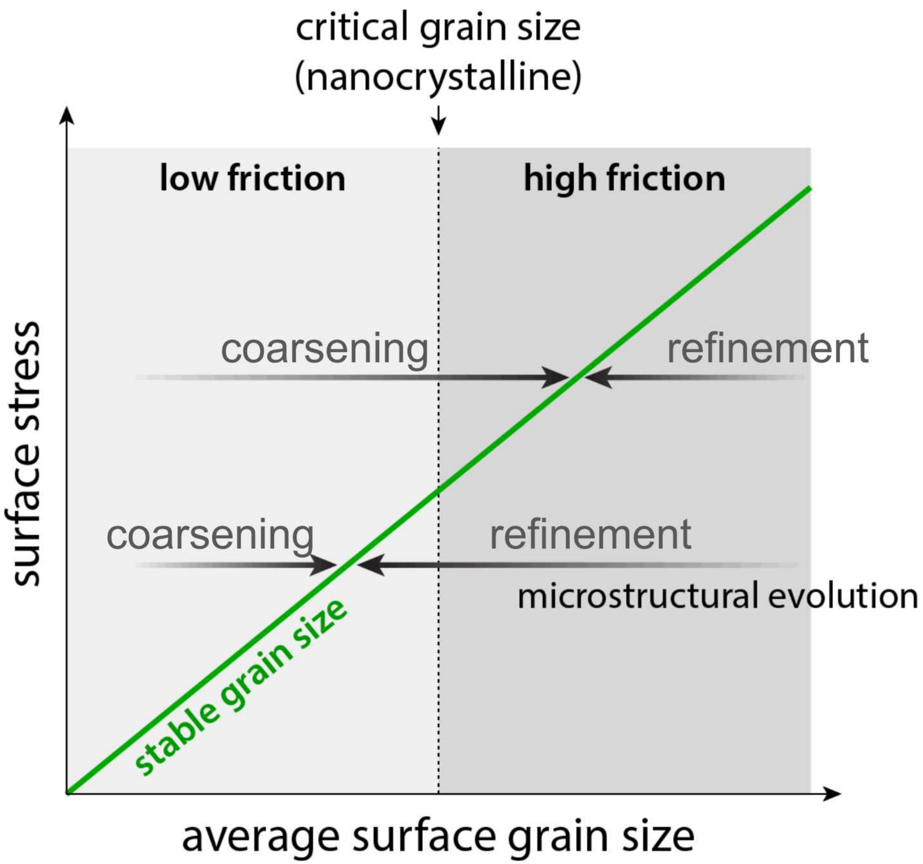


"Crystals are like people, it's the defects that make them interesting."
- C. J. Humphreys

Crystal Structure of Metals (A Primer)



Grain size evolution is a competition (growth/refinement)



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

"It is better to be vaguely right than exactly wrong."
--Carveth Read

- Effective refinement from recrystallization (Zener & Holloman, 1944; McQueen et al., 1967)
- Known in rocks and ice cores (Derby et al., 1992)
- Recently extended to metals under severe plastic deformation (Pougis et al., 2014)

N. Argibay, et al., ***J. Mat. Sci. (2017)***