Amorphous Dielectric Coatings

M.M. Fejer

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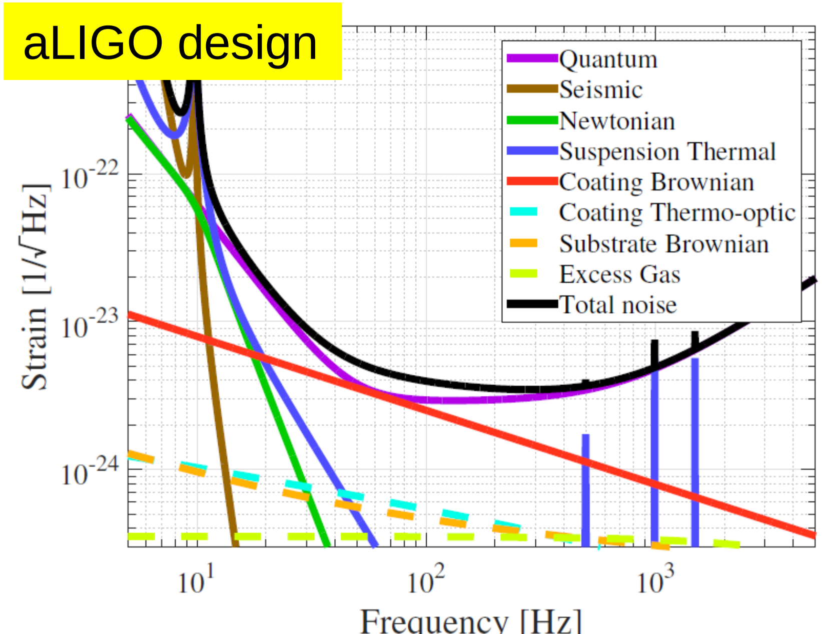
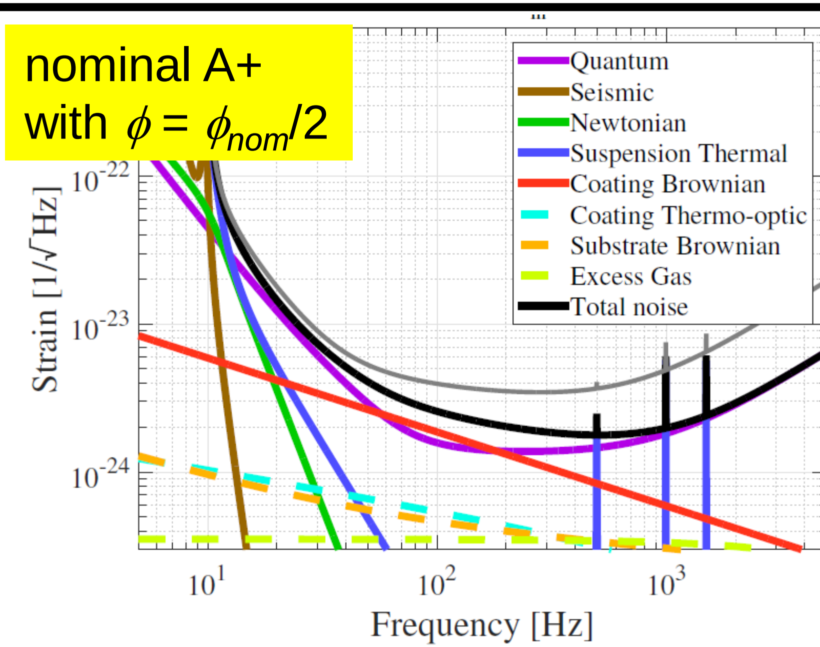
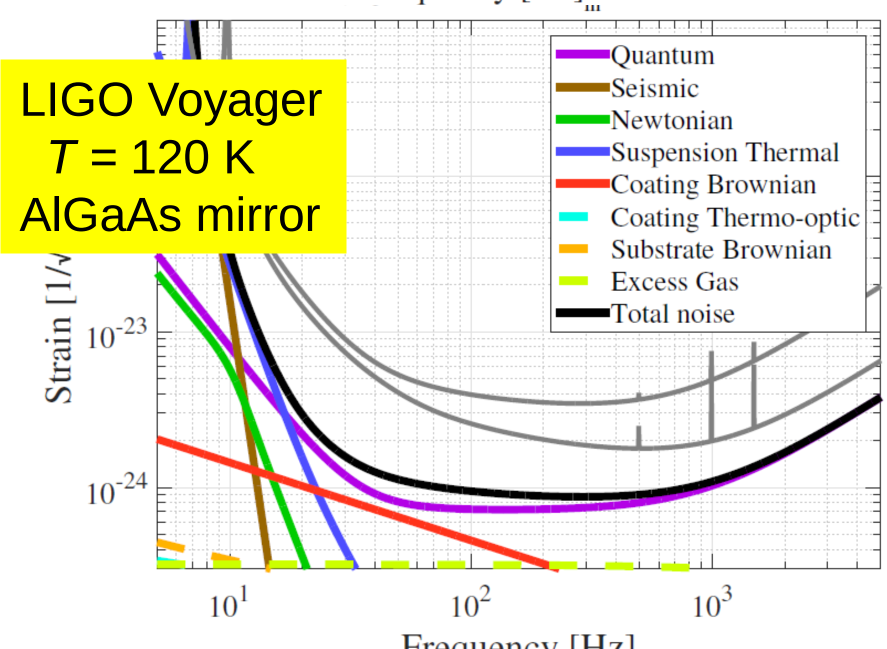
GWADW 2016

Reduced Elastic Loss in LIGO Mid-Band







coating elastic loss

coating thickness

$$S_x(f,T) \approx \frac{2k_BT}{\pi^2 f}\frac{d}{w^2Y}\phi\left(\frac{Y'}{Y}+\frac{Y}{Y'}\right)$$

beam radius

$$\phi_{\mathrm{TiO_2:Ta_2O_5}} = 2\times10^{-4}$$
$$\phi_{\mathrm{SiO_2}} = 4\times10^{-5}$$
$$\phi_{\mathrm{AlGaAs}} = 2.4\times10^{-5}$$

Plots from LIGO-T15TBI-v1, I.S. White Paper

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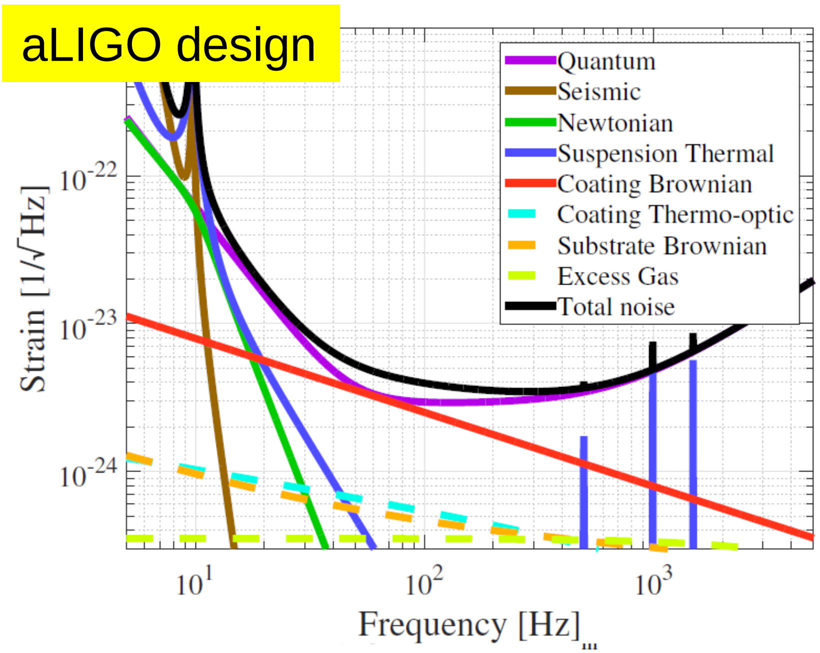

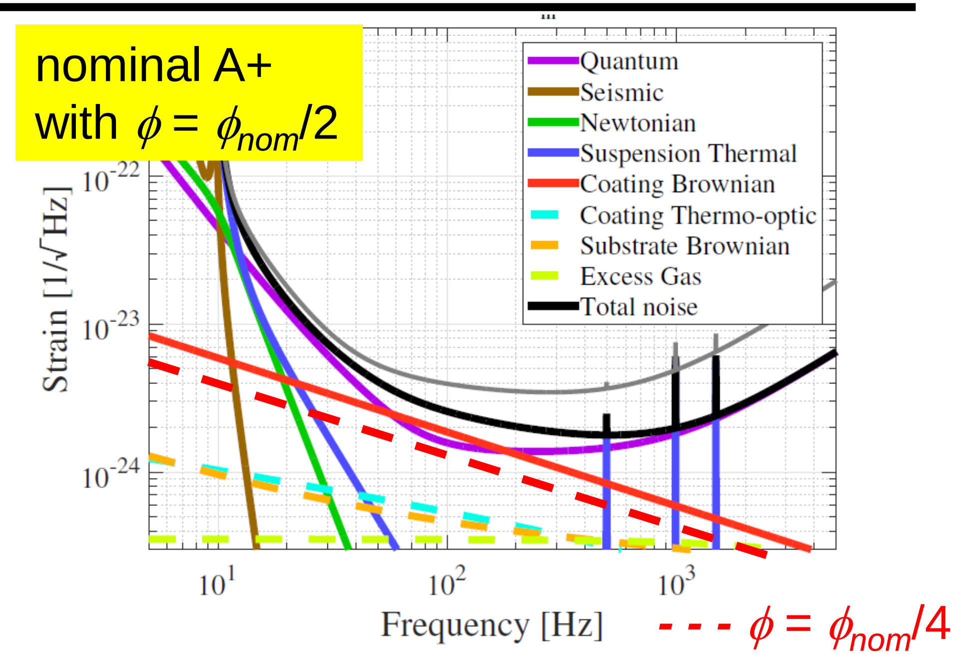

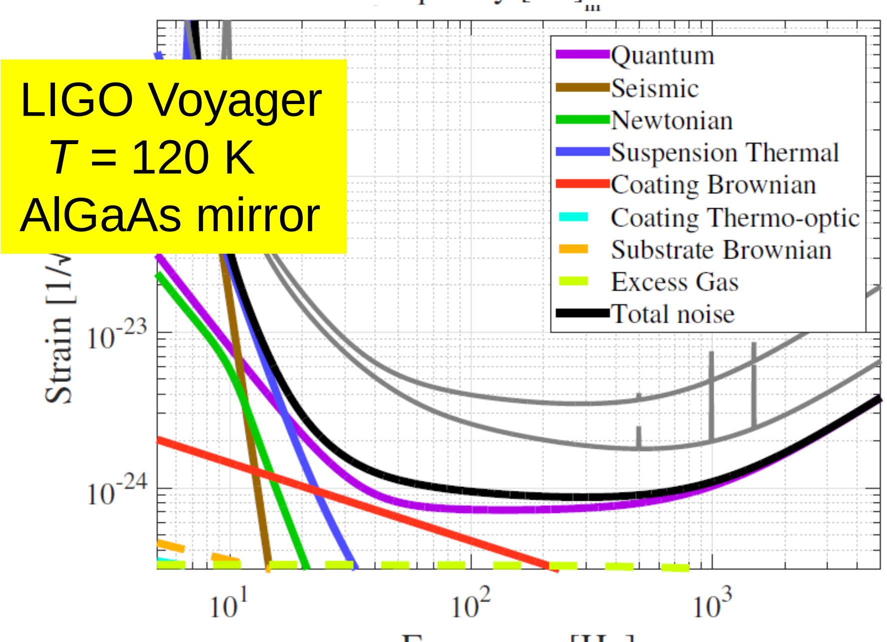

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Plausible Goals

- A+ LIGO
  - room temperature $\phi < 1x10^{-4}$

- LIGO Voyager (cryogenic)
  - 120 K $\phi < 2x10^{-5}$

- While maintaining optical properties over 34 cm diameter

  at 1.06 μm for A+-LIGO

  at ~1.5 - 2 μm for cryo LIGO Voyager
  - optical absorption ~0.5 ppm
  - micro-roughness scatter ~1 ppm

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- Useful topic for discussion:
  - tighten up these numbers and required timeline

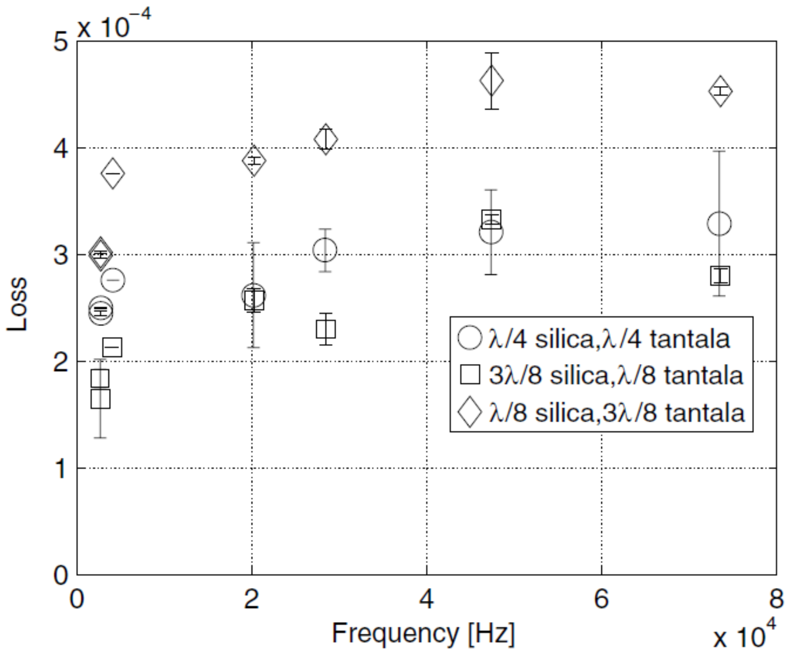
Outline

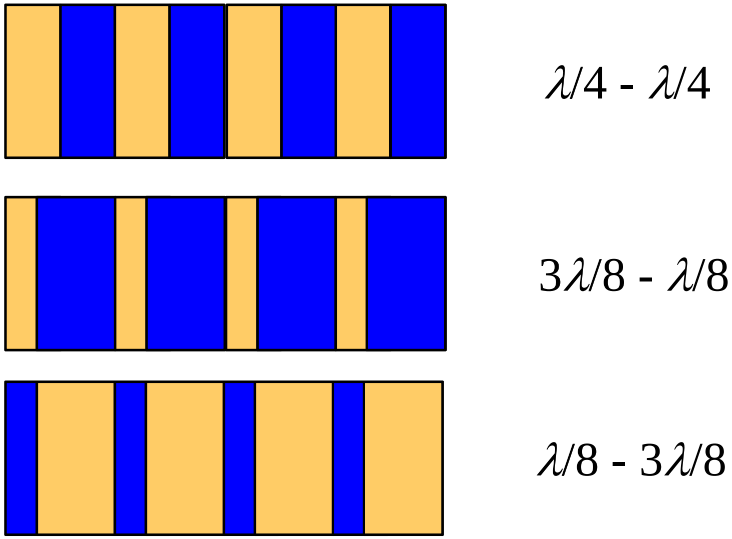
- Quick sketch of what we (think we) know
- Two-level system (TLS) model for internal friction
- Theoretical guidance
  - molecular dynamics
- Amorphous silicon
  - potential material for A+^{n} and LIGO Voyager
  - model system for ultra-stable glass
- More theory
  - “Ultra-stable glasses” from high-temperature deposition
- Structural characterization
- Some thoughts on strategy going forward

General Observations About Coating Elastic Loss

Volume and Interface Effects

- Volume rather than interface losses dominate in tantala/silica mirror





D. Crooks, Class. Quantum Grav. 23 (2006) 4953–4965

– current values: Ti:tantala ~5x lossier than silica

– thickness independent

some suggestion of thickness dependence in a-Si

D.R. Queen, F. Hellman, et al, *Journal of Non-Crystalline Solids* **426** 19–24 (2015)

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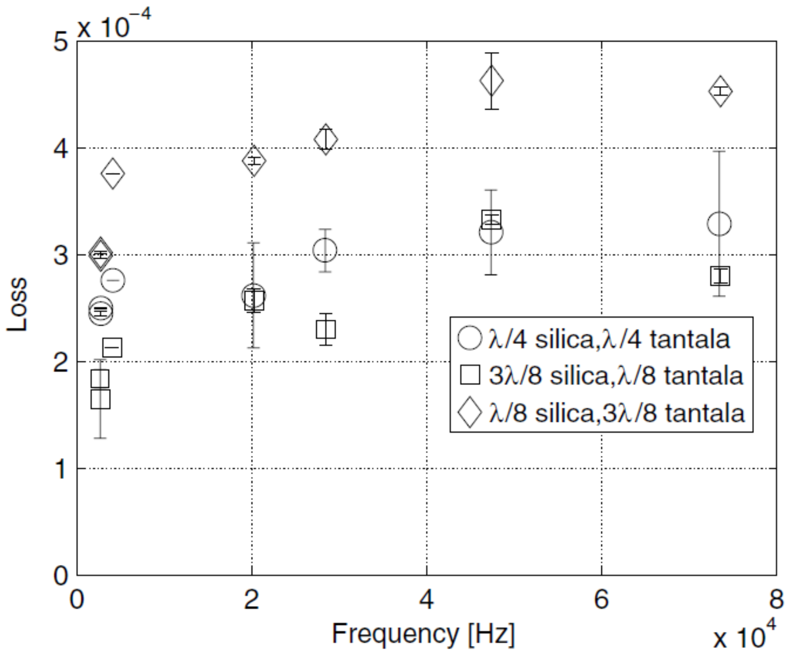

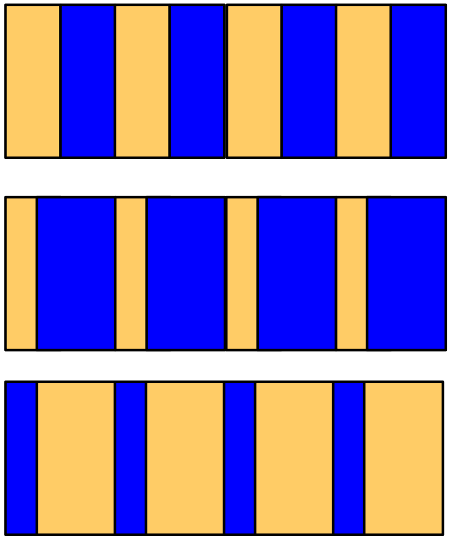

$\lambda/4$ - $\lambda/4$

$3\lambda/8$ - $\lambda/8$

$\lambda/8$ - $3\lambda/8$

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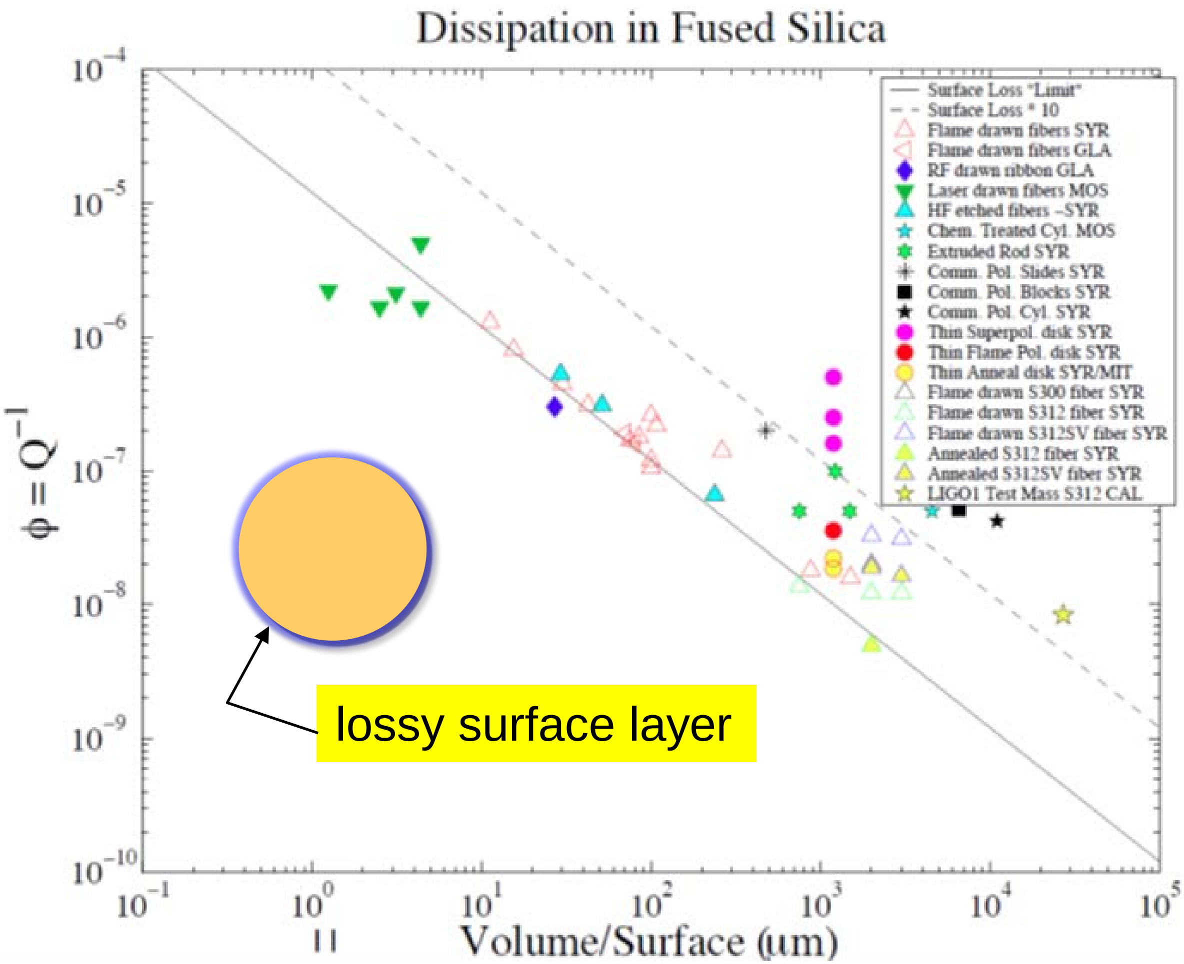
health warning:
new thoughts from Geppo
talk Wednesday

Surface and Volume Loss in Bulk Silica

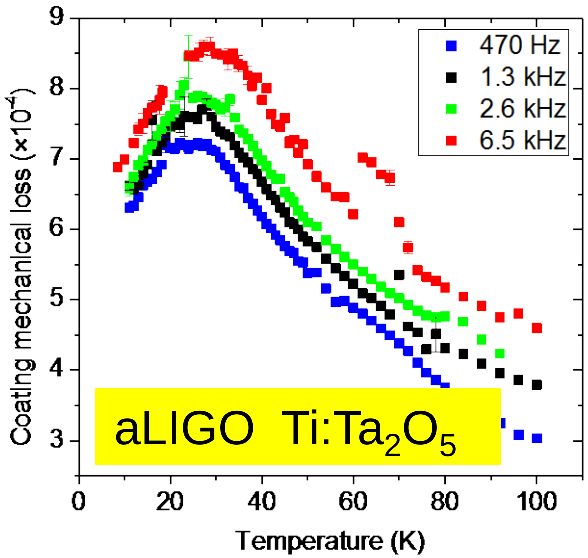
- Bulk and surface loss
- Decrease with:
  - annealing temp.
    up to ~900°C
  - annealing time
- Rayleigh scattering also decreases with annealing
  - $\Rightarrow$ density fluctuations reduced

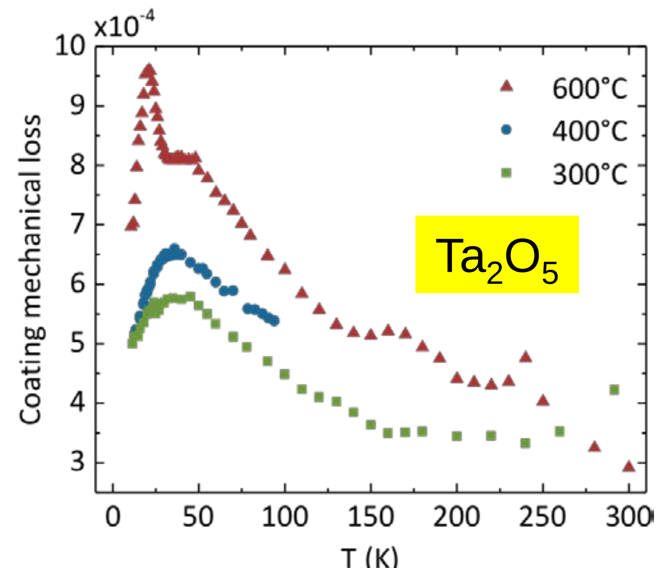


S. Penn, LIGO-G1300776 (2013)

Temperature and Frequency Dependence

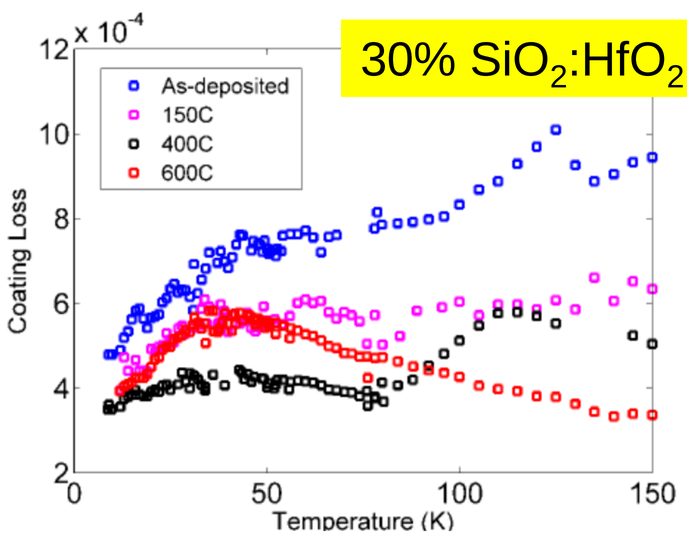
- Typical behavior vs T and f

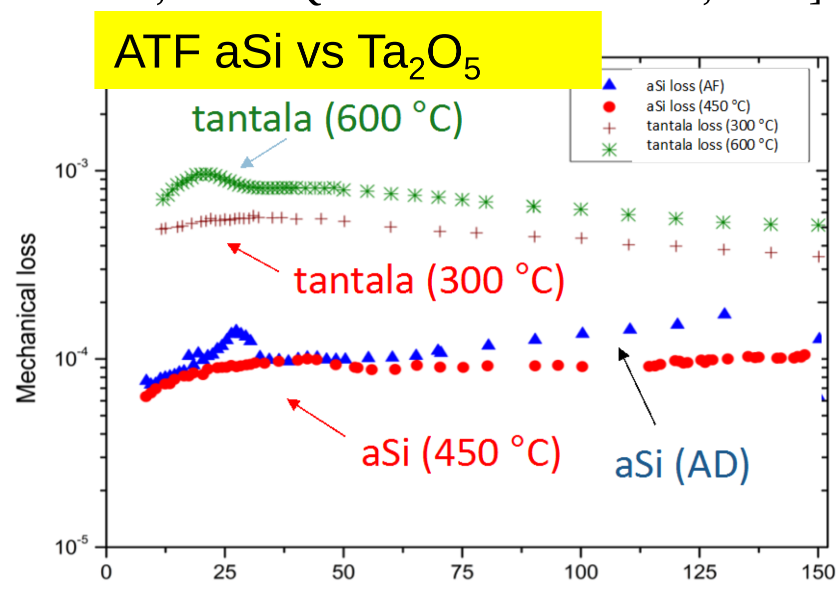




[I W Martin et al, Class. Quant. Grav. 27 225020, 2010]

- Other materials behave differently

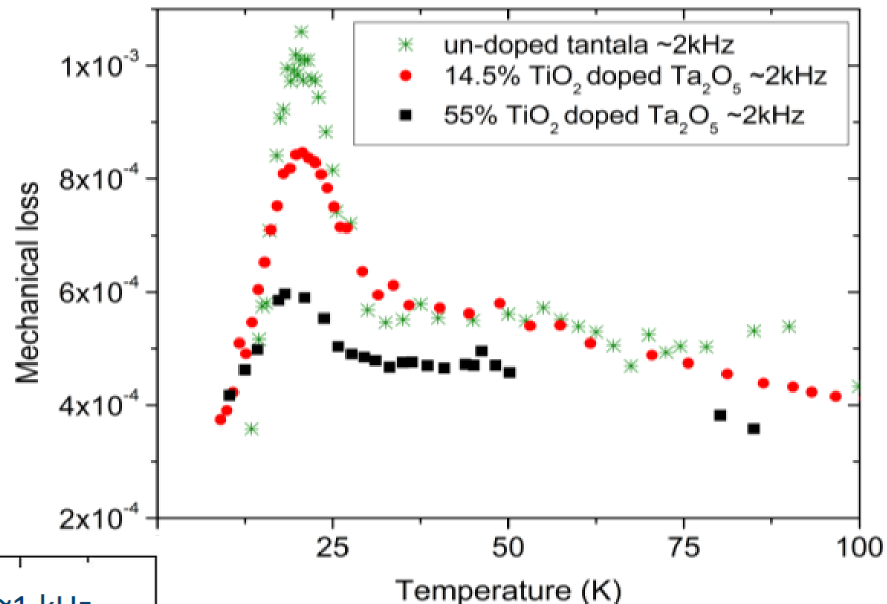




I. Martin et al., Glasgow

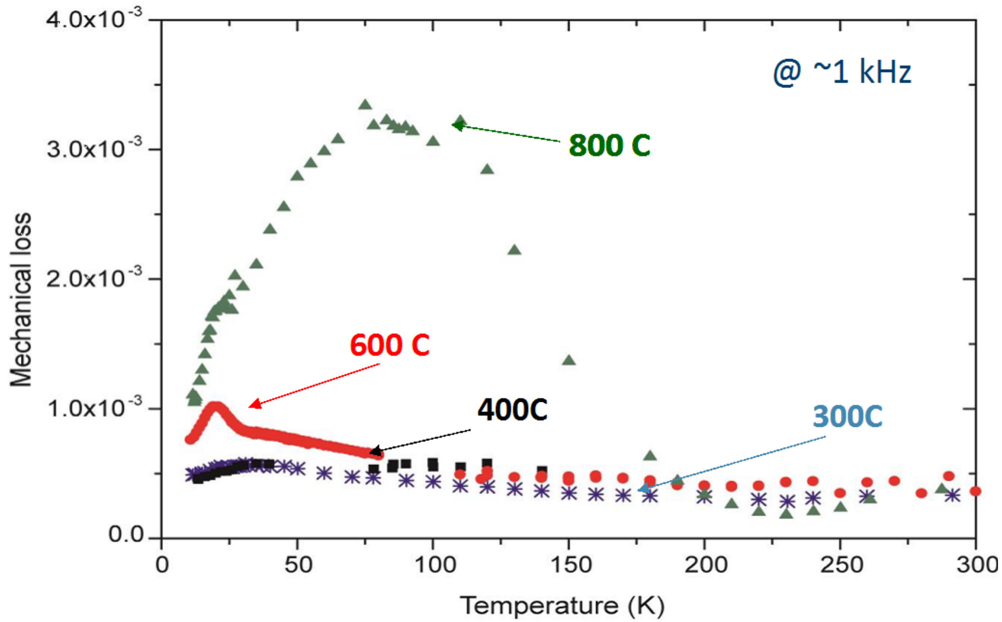
Doping and Annealing Alter Dissipation

- Loss modified by dopants
  - TiO_2 doping
    reduces losses in Ta_2O_5



P. Murray et al, U. Glasgow
- (Unpublished)

- Annealing modifies loss spectra



  - can improve at some temperatures while worsened at others

How to Find Better Amorphous Oxides?

- Don't bother: use crystalline coatings

- Random search through ternary oxides?
  - thousands of host/dopant combinations
  - thermal processing another degree of freedom

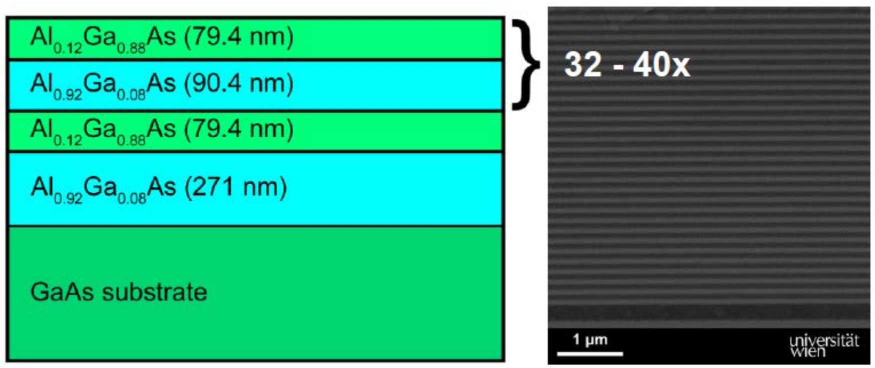
- *Ars longa, vita brevis* (Art is long, life is short)
  - Edisonian methods used to date may not scale gracefully
    - limited time and $$ available

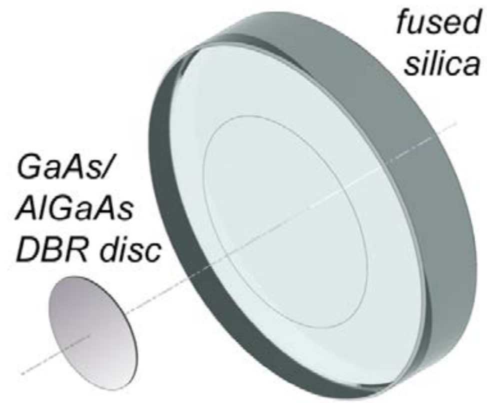
- Understanding underlying mechanisms useful to guide experiment
  - two-level systems (TLS)
  - molecular dynamics modelling
  - atomic structure characterization

How to Eliminate TLS?: Crystalline Coatings

- GaAs/AlGaAs transferred to mirror substrate

  $\phi \sim 2 \times 10^{-5}$, $\alpha < 1$ ppm



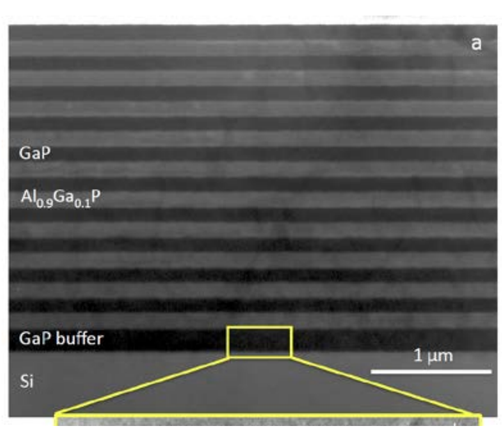


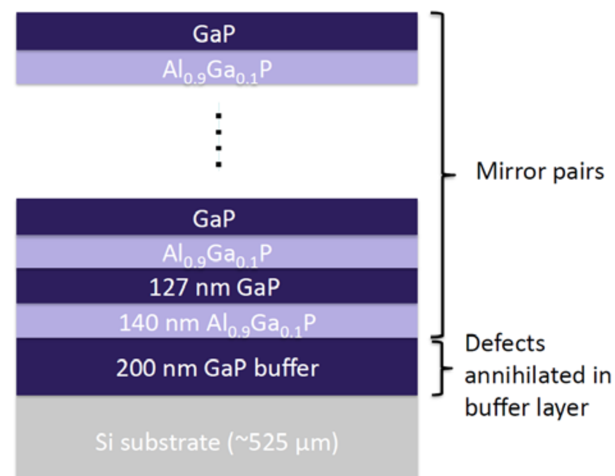
Garrett Cole talk, Friday 2:00

G. Cole, LIGO-G1401152

- GaP/AlGaP epitaxy on Si mirror substrate

  $\phi < 1 \times 10^{-4}$, $\alpha = ??$





A. Lin, LIGO-G1200135

How to Reduce TLS: Find Better Amorphous Oxides?

- Don’t bother: use crystalline coatings

- Random search through ternary oxides?
  - thousands of host/dopant combinations
  - thermal processing adds another degree of freedom

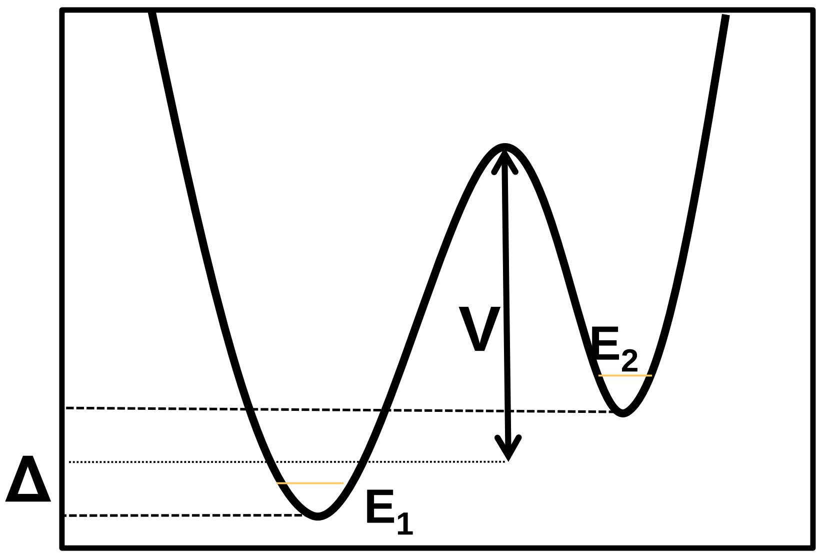
- *Ars longa, vita brevis*
  - Edisonian methods used to date may not scale gracefully

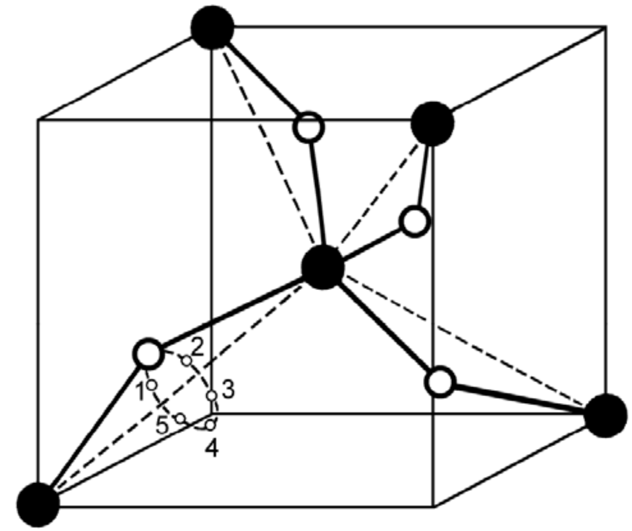
    limited time and $$ available

- Understanding underlying mechanisms useful to guide experiment
  - two-level systems (TLS)
  - molecular dynamics modelling
  - “ultra-stable” glasses
  - atomic structure characterization

Low-frequency losses in amorphous dielectrics

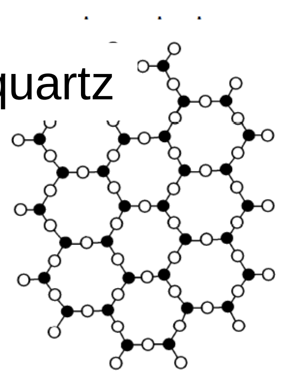
- Conventionally associated with low energy excitations (LEEs)
  - conceptualized as two-level systems (TLS)





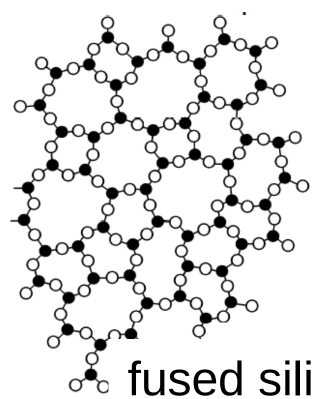
Oversimple picture: bond flopping

crystal quartz



(● = Si, ○ = O).

A)



fused silica

Distribution of TLS in silica due to disordered structure

figures from B.S. Lunin monograph

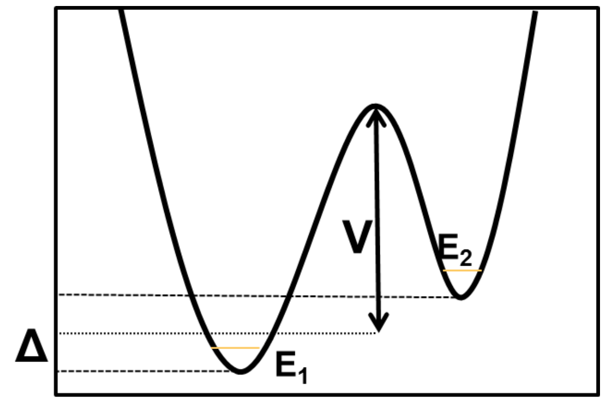
Elastic Dissipation by TLS

- Above ~5 K, thermally activated rather than quantum tunneling
  - for a single type of TLS:

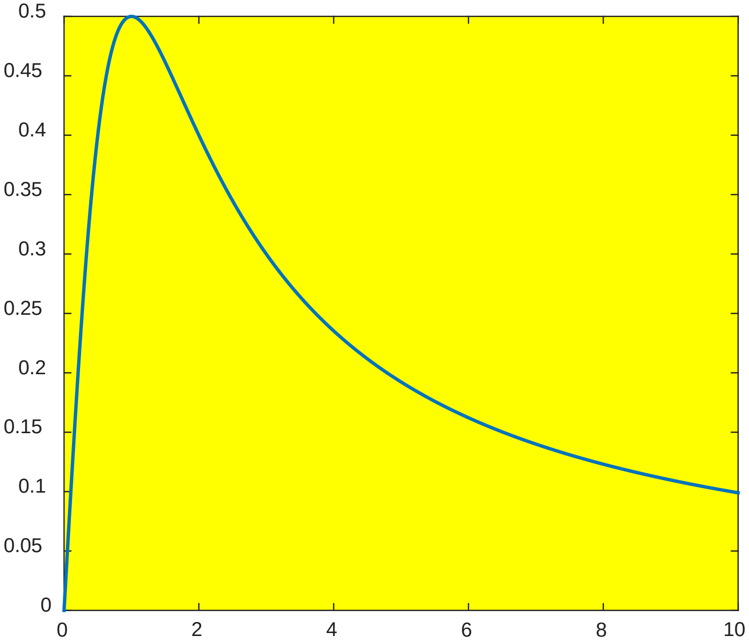
$$Q^{-1} = N\frac{\gamma^2}{Y\,kT}\frac{\omega\tau}{1+\omega^2\tau^2}\mathrm{sech}^2\left(\frac{\Delta}{kT}\right)$$

$$\tau = \tau_0\,\mathrm{sech}\left(\frac{\Delta}{2kT}\right)e^{V/kT}$$

ω: acoustic frequency

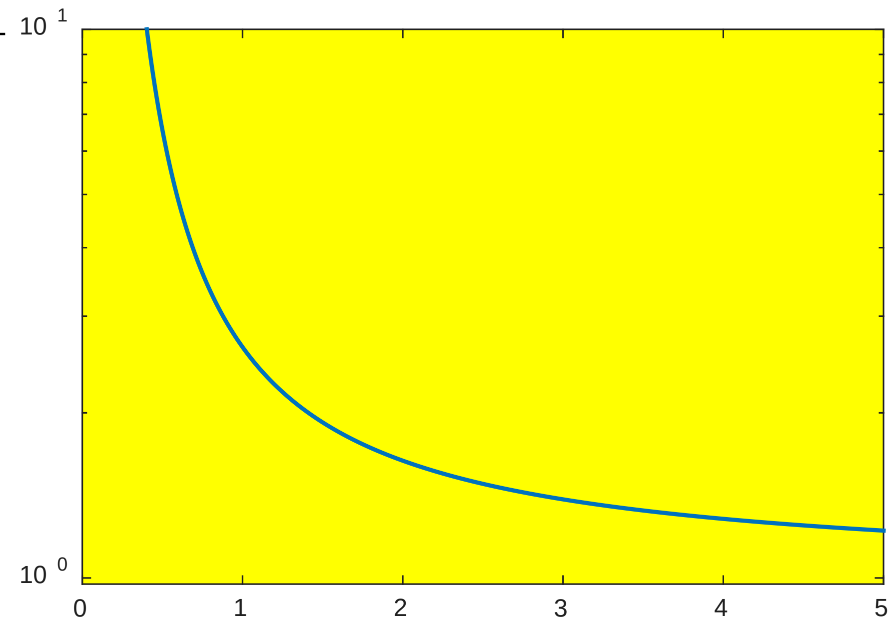


$$\frac{\omega\tau}{1+\omega^2\tau^2}$$



$\omega\tau$

$$\frac{\tau}{\tau_0}$$



$kT\,/\,V$

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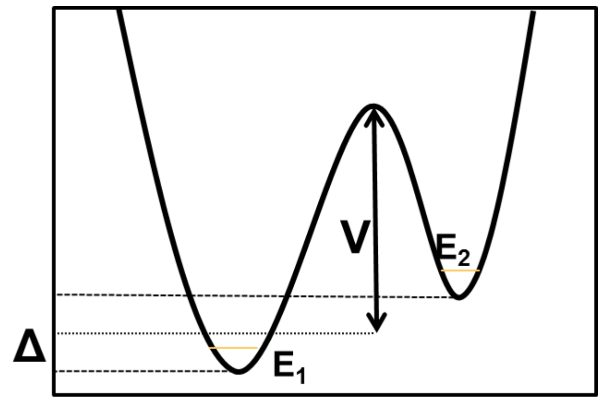

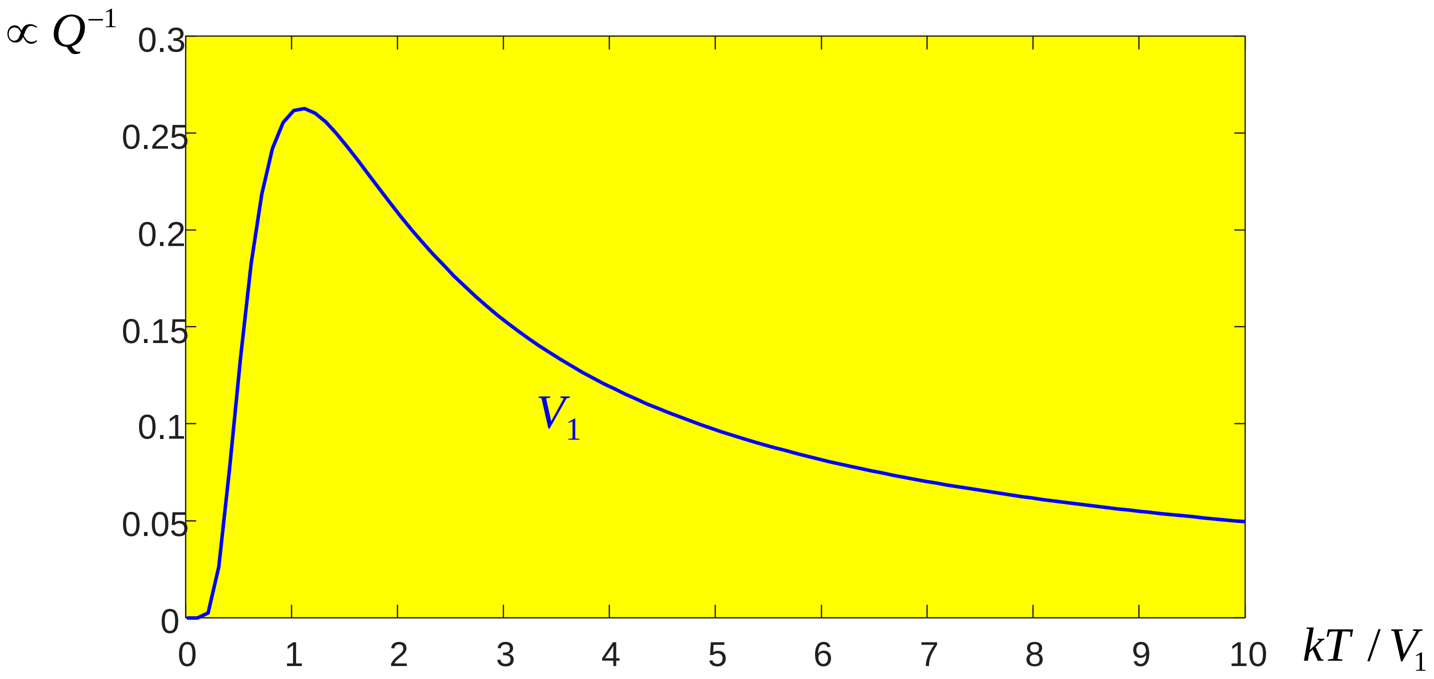

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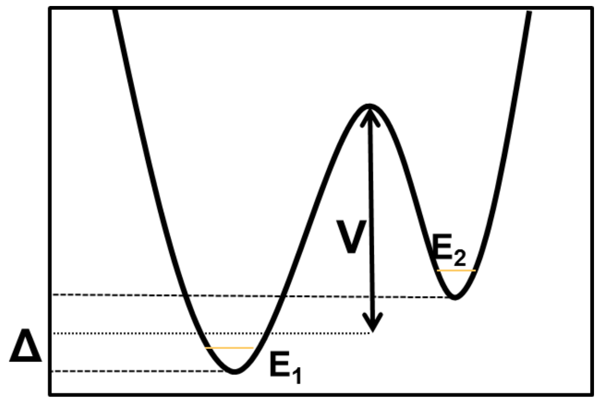
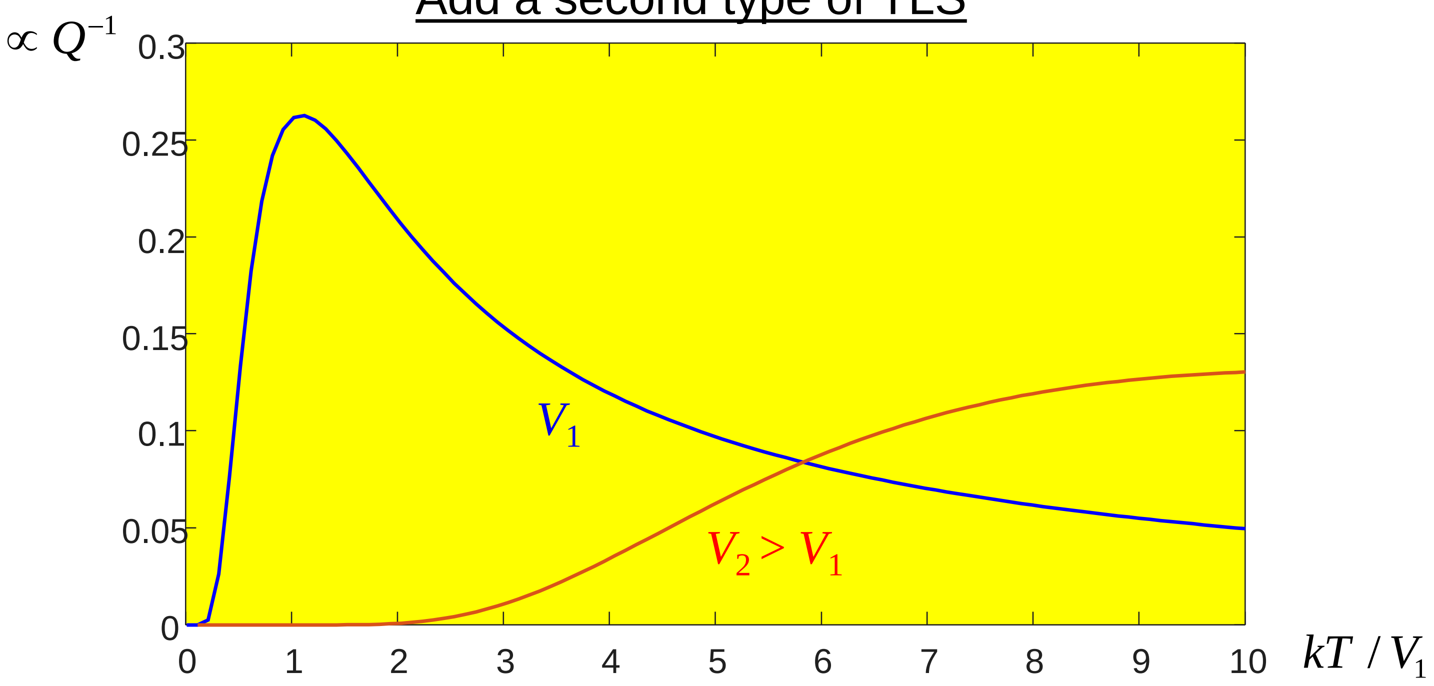


Add a second type of TLS



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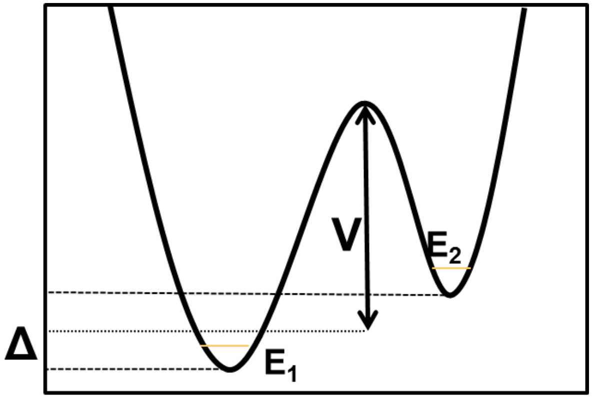

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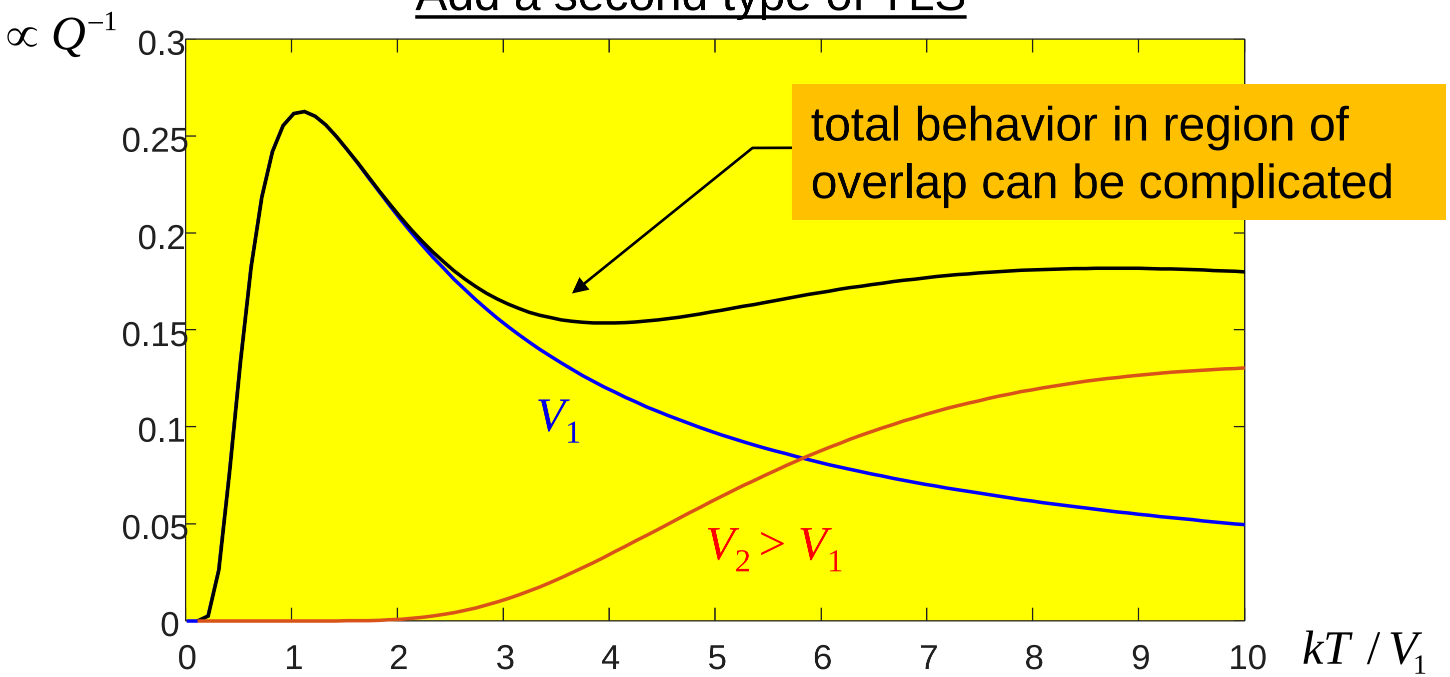
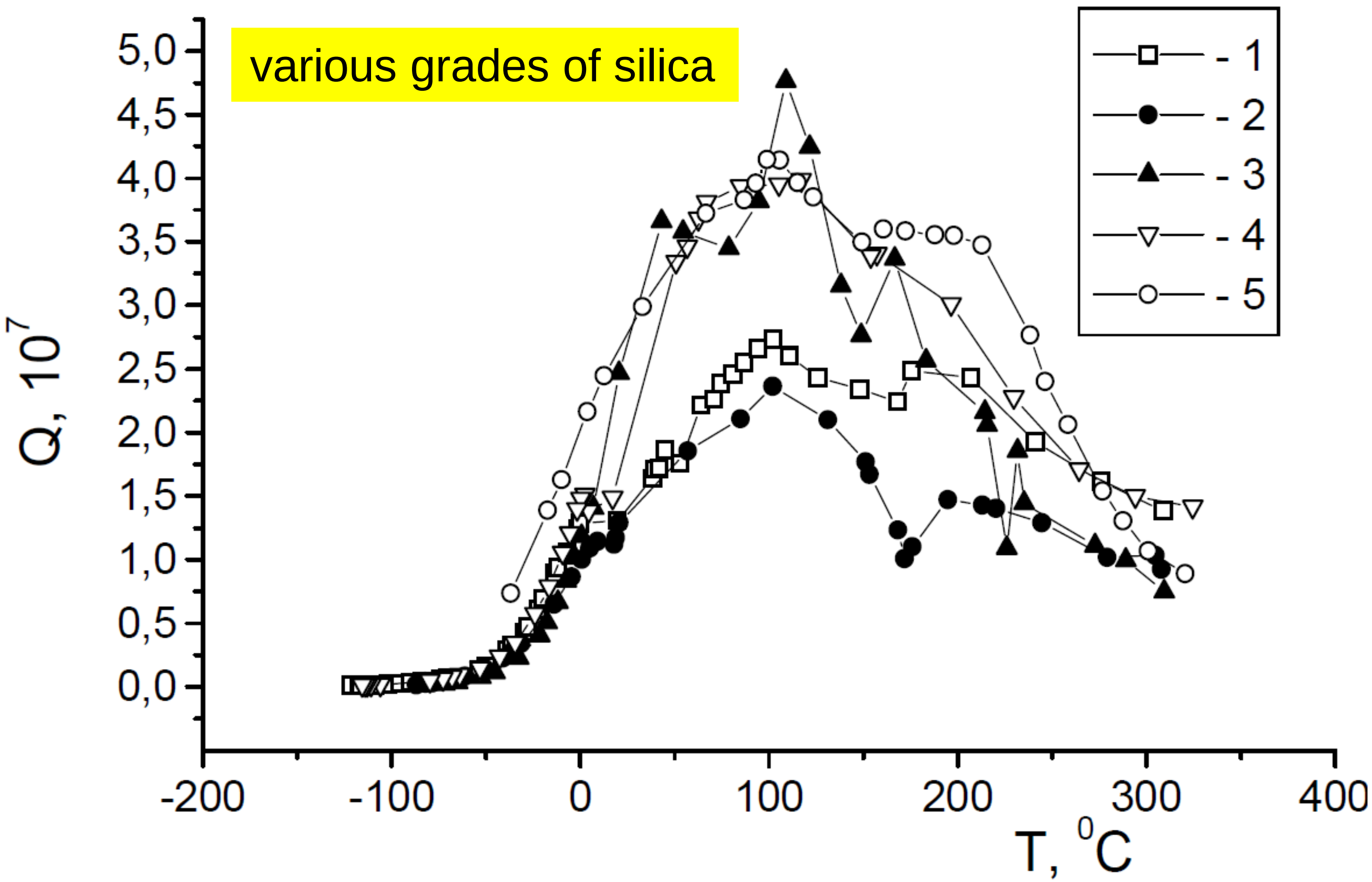


Evidence of Such Behavior in Silica



from Lunin monograph

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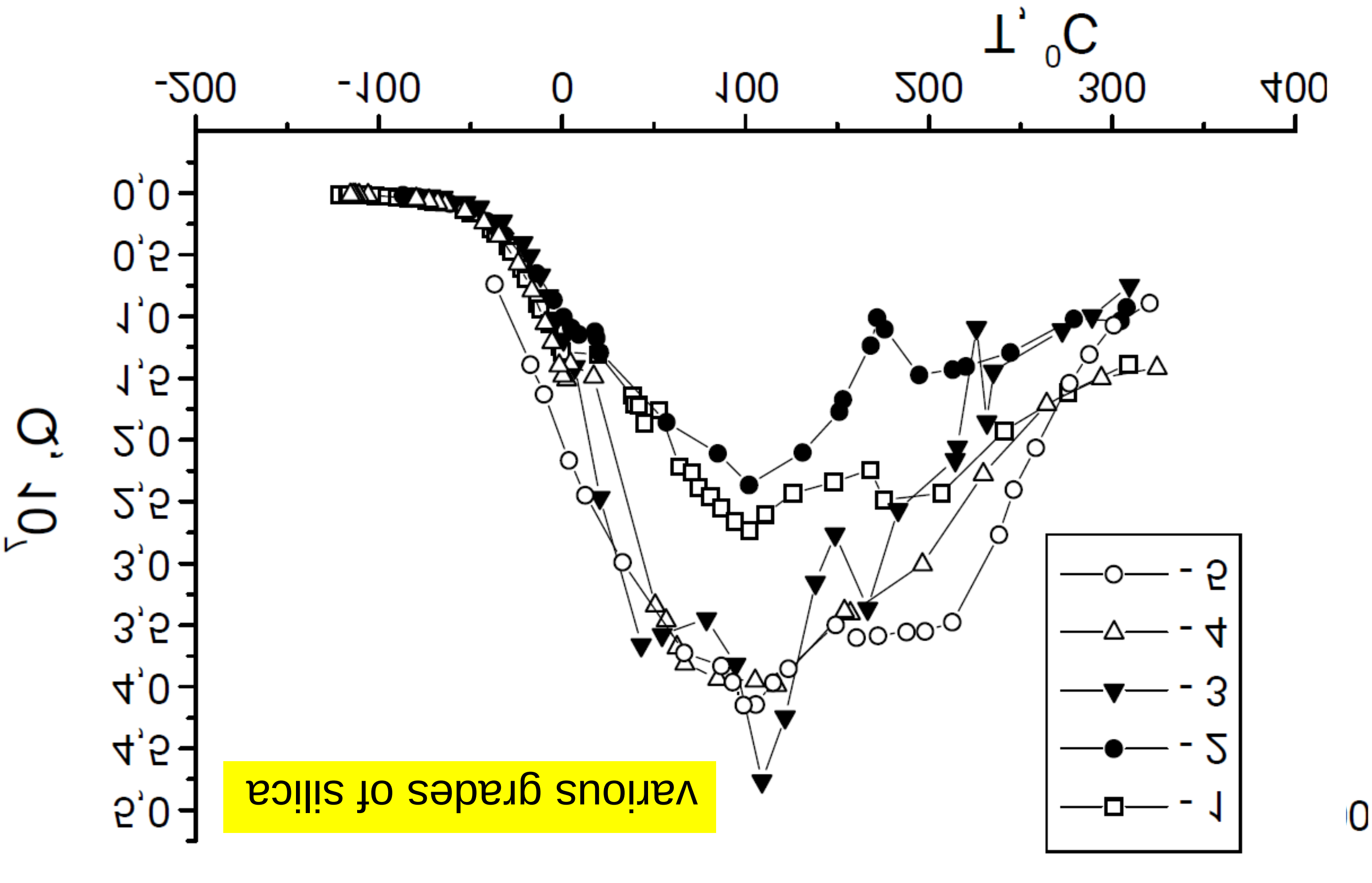


from Lunin monograph

Elastic Dissipation by TLS

- Real materials have distribution of TLS

Gilroy, K. S. and Phillips, W. A., Philos Mag. B, **38,** 735 (1981)

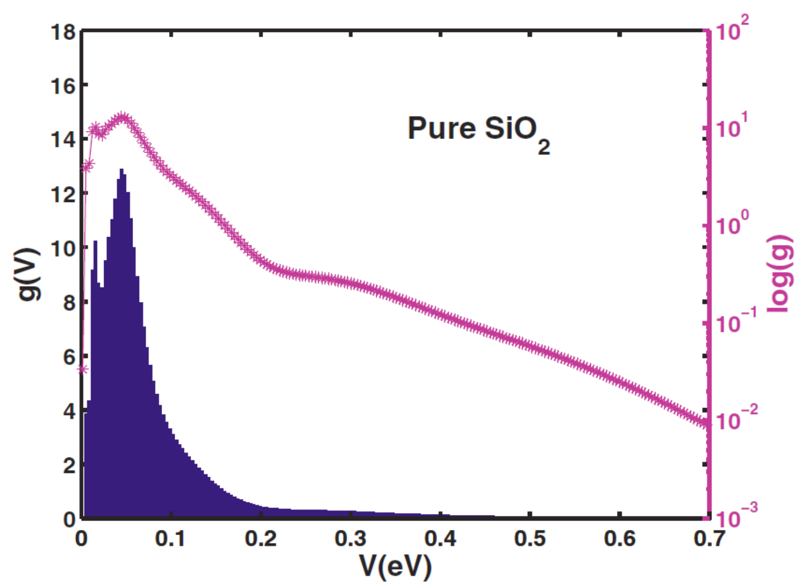
- Dissipation is weighted average of their contributions
  - smears out attenuation function

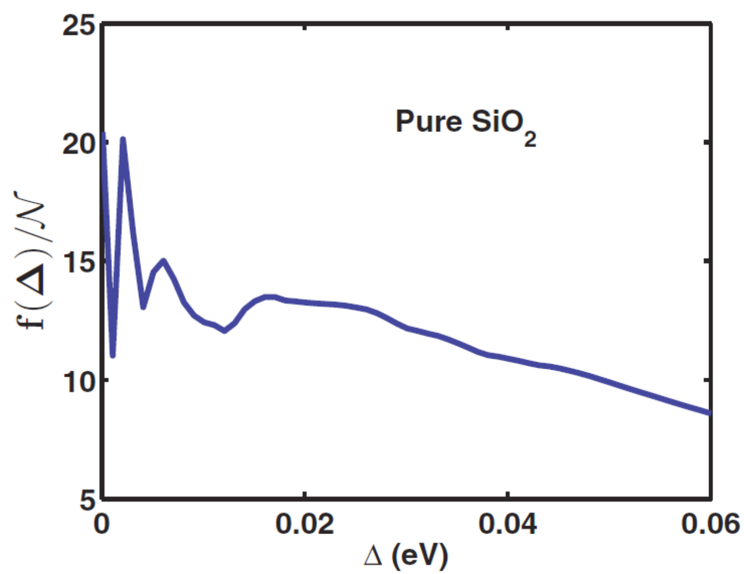
$$Q^{-1} = \frac{\gamma^2}{Y\,kT} \iint \frac{\omega\tau}{1+\omega^2\tau^2} \operatorname{sech}^2\left(\frac{\Delta}{kT}\right) g(V)\, f(\Delta)\, dV\, d\Delta$$

barrier height distribution

asymmetry distribution

$$\tau = \tau_0 \operatorname{sech}\left(\frac{\Delta}{2kT}\right) e^{V/kT}$$



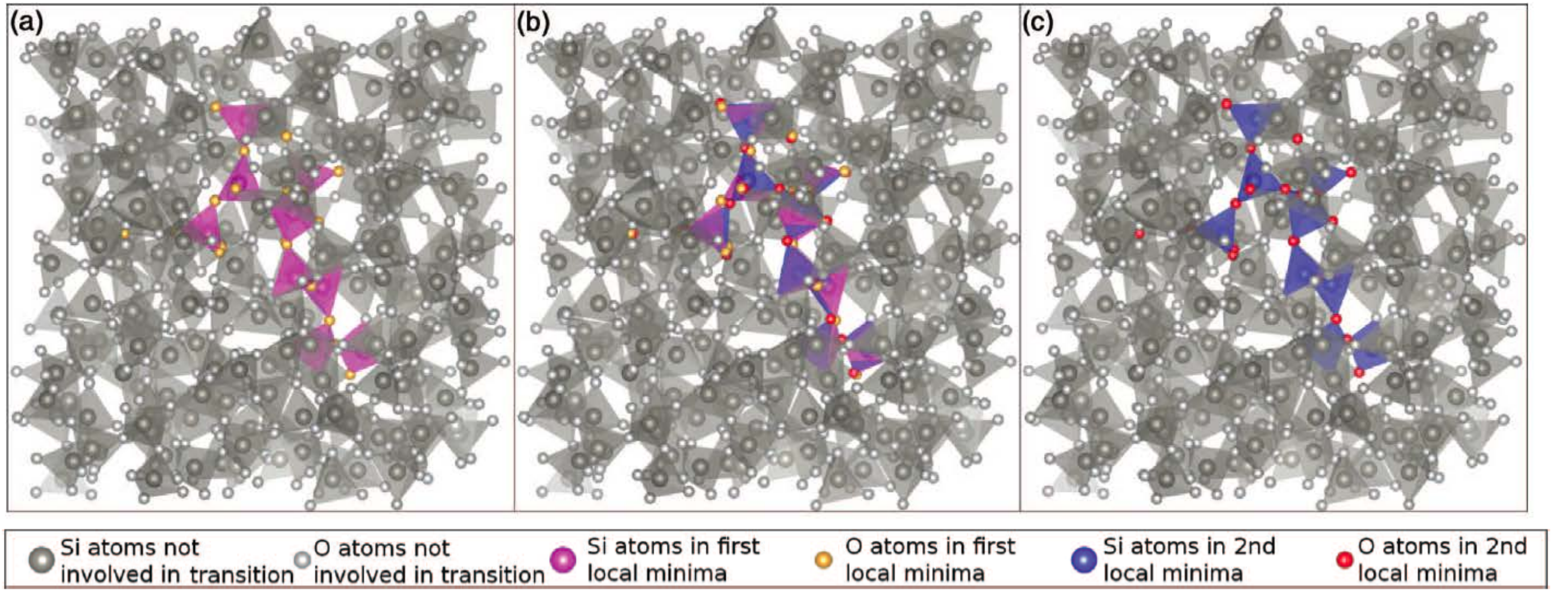


Calculated in R. Hamdan, J.P. Trinastic, and H. P. Cheng, J. Chem.Phys. **141**, 054501 (2014)

Theoretical Guidance: Molecular Dynamics

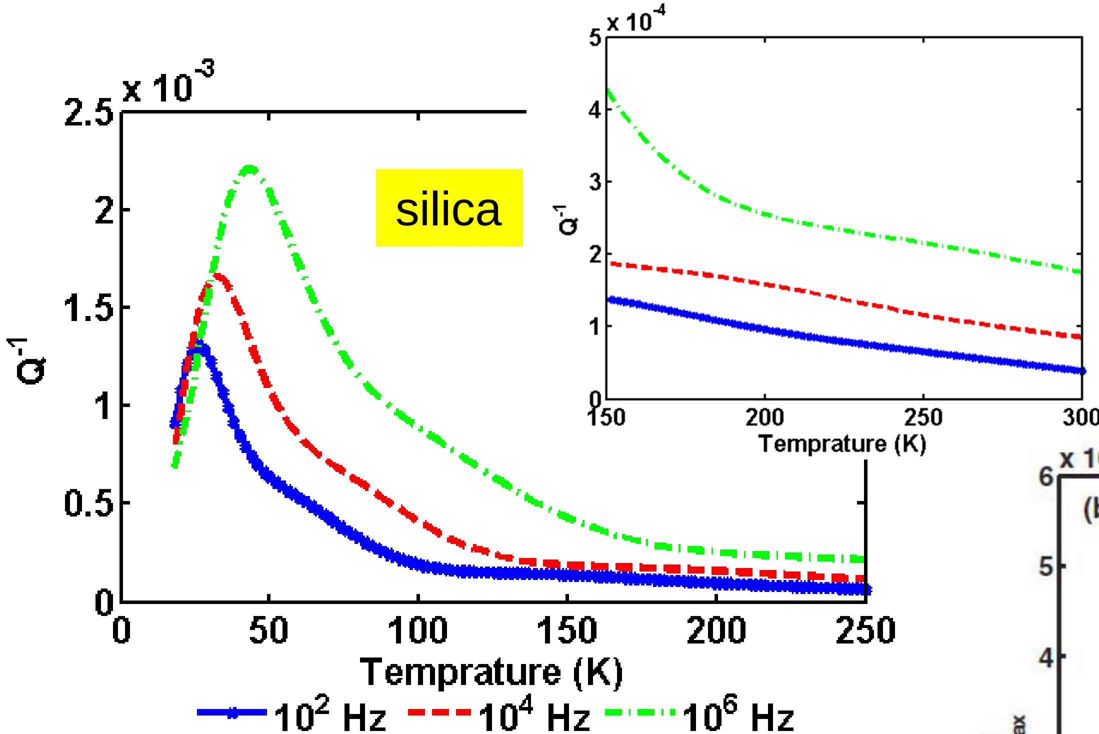
- Molecular dynamics calculations for amorphous materials
  - provide insight into dissipation mechanisms
  - can suggest promising material combinations
  - Chris Billman talk at 10:00 Tuesday

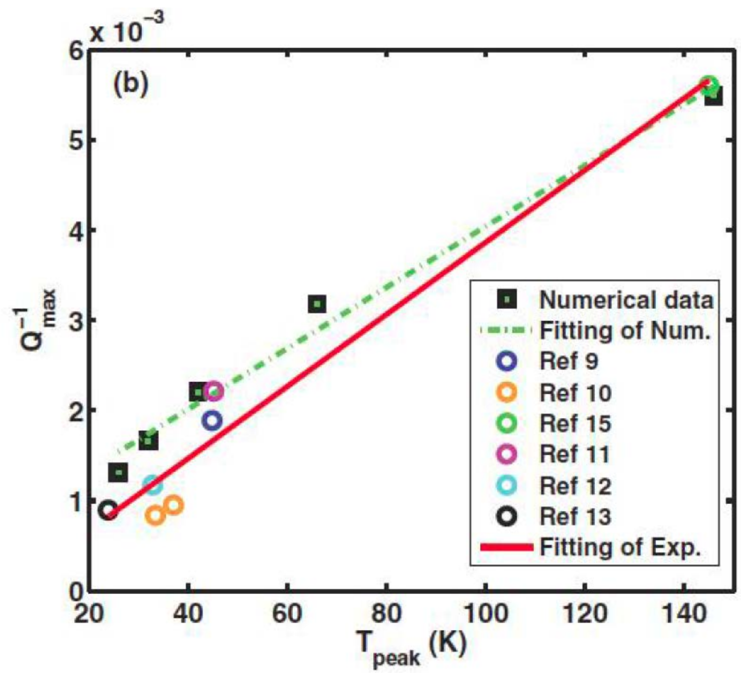
- Some observations:
  - TLS involves dozens of atoms in nm-scale configurations



Theoretical Guidance: Molecular Dynamics

- Some observations:
  - some theoretical trends tie up with experiment



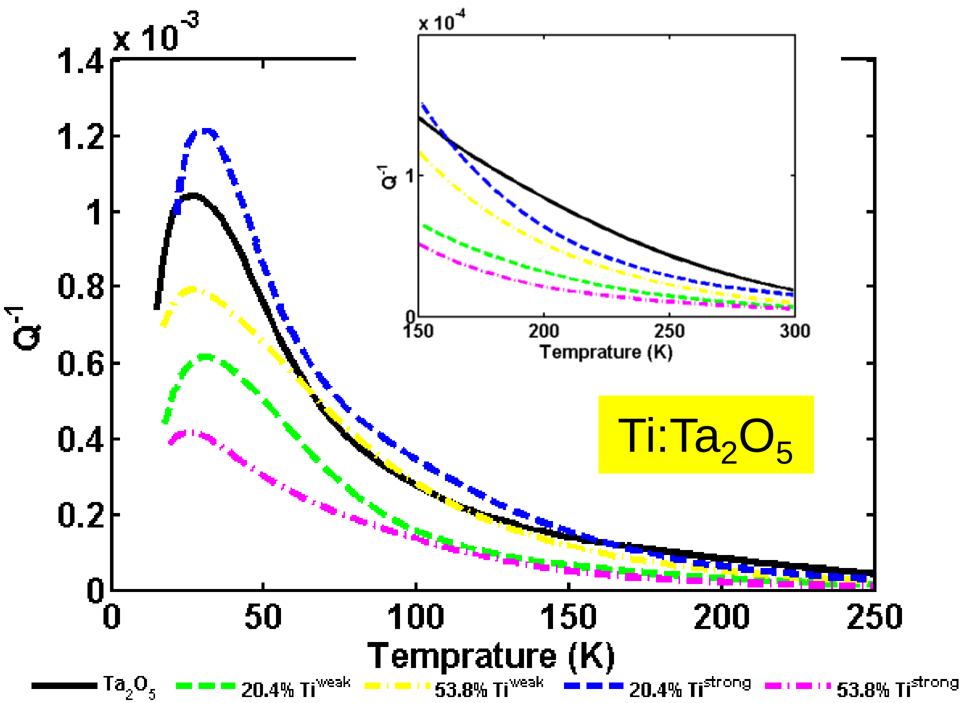


R. Hamdan, H. Cheng, *J. Chem. Phys.* **141**, 054501 (2014)

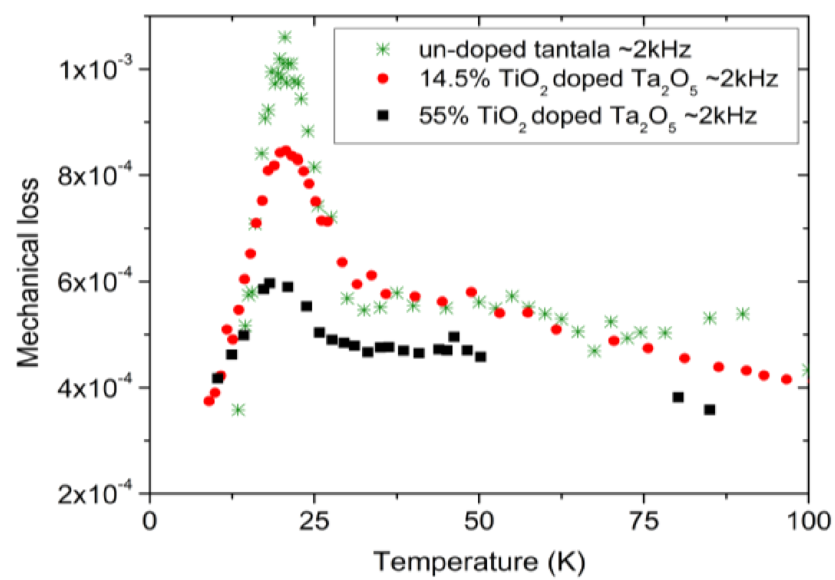
Theoretical Guidance: Molecular Dynamics

- Some observations:
  - some theoretical trends tie up with experiment

    decrease in loss with titania dopoing in tantala
-



JP Trinastic, *PRB* **93**, 014105 (2016)



P. Murray et al, U. Glasgow

- (Unpublished)

Theoretical Guidance: Molecular Dynamics

- Some issues:
  - absolute values not in close agreement with exp't
  - agreement worse at higher temperatures than cryogenic

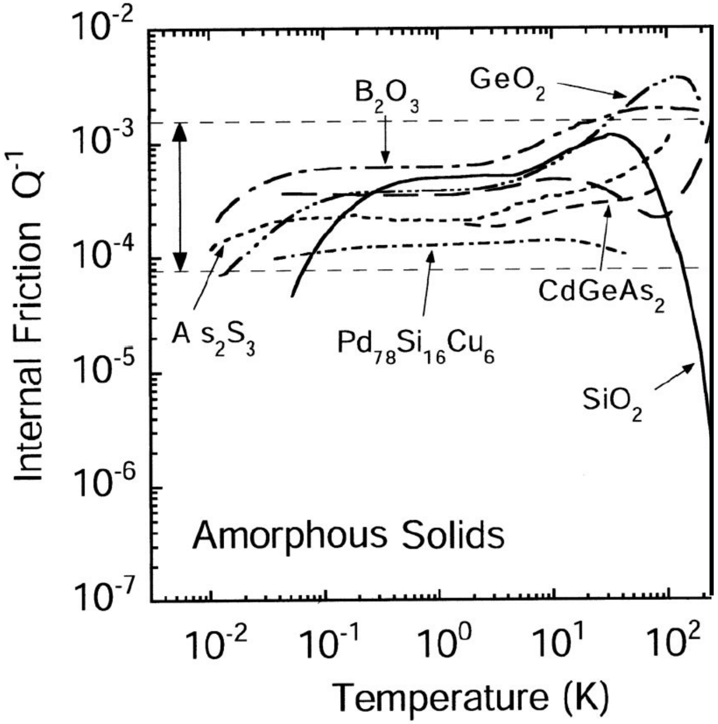
- Possible sources:
  - surface effects
  - higher energy TLS
  - ??

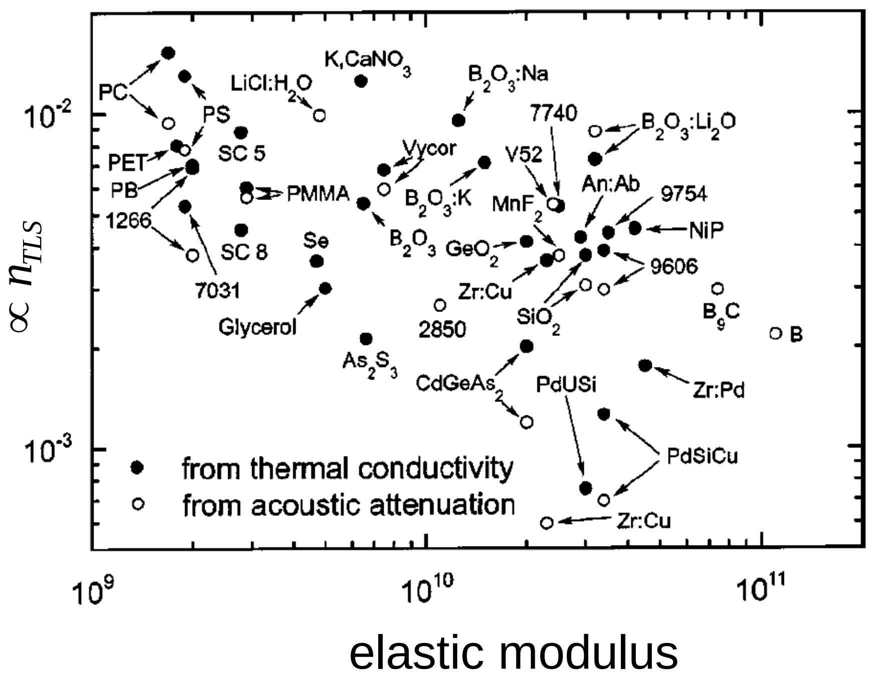
- Correct trends already suggest potentially interesting materials
  - e.g. ZrO_2:Ta_2O_5

Chris Billman talk at 10:00 Tuesday

How to Reduce TLS? Synthesis and Annealing

- Universality of TLS in amorphous materials recently studied





- Has led to studies of elevated vapor deposition temperatures
  – very promising results

R.O. Pohl, X. Liu, and E. Thompson, *Rev. Mod. Phys.* **74**, 991 (2002)

Several Properties $\propto$ concentration of TLS

- At low temperatures several properties $\propto n_{TLS}$

- Internal friction (in tunneling regime)

$$\phi \propto \bar{P} \propto n_{TLS}$$

spectral density of TLS

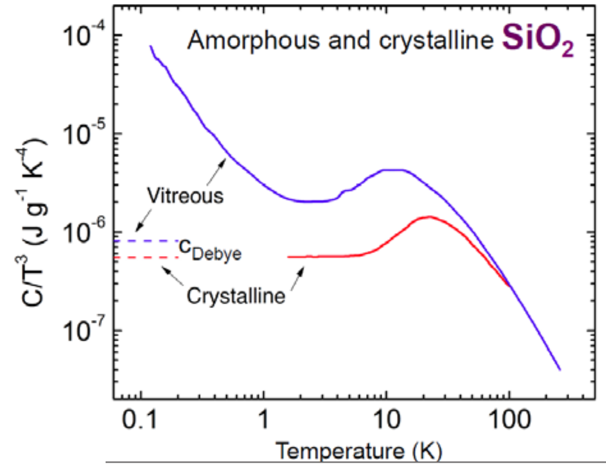
- Specific heat vs temperature T

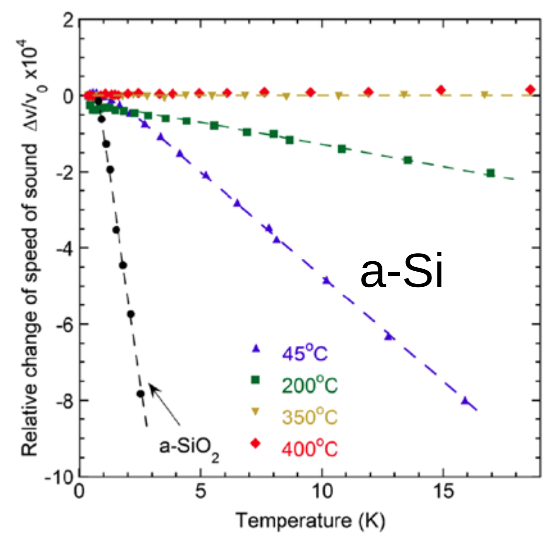
$$C = c_1 T + c_3 T^3$$

phonons

$$c_1 \propto n_{TLS}$$

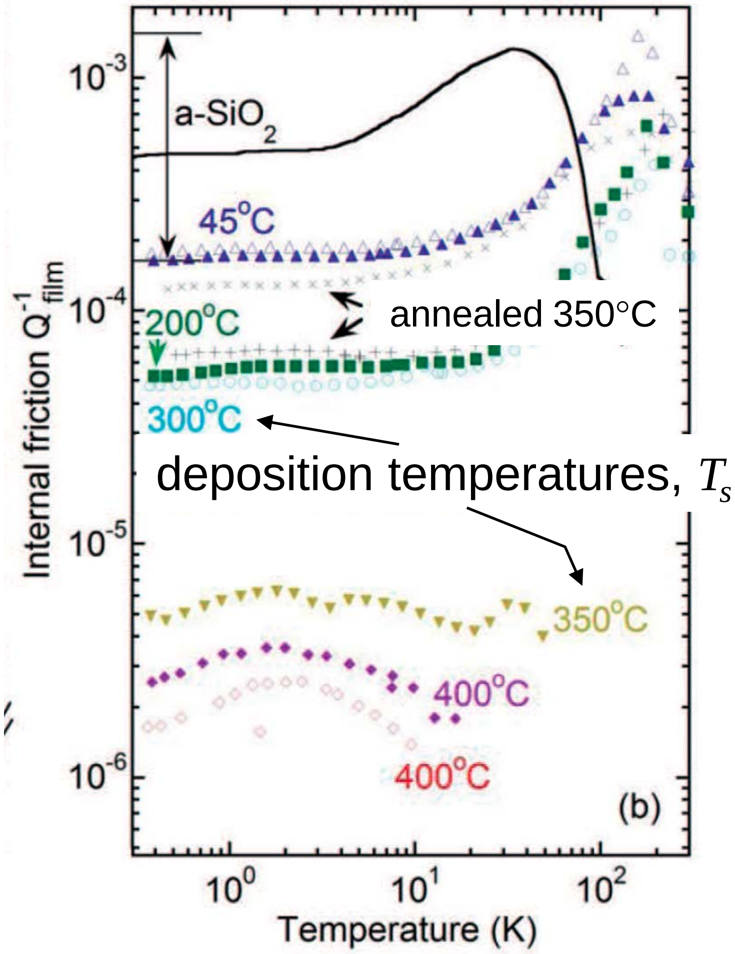
- Change in sound velocity vs T





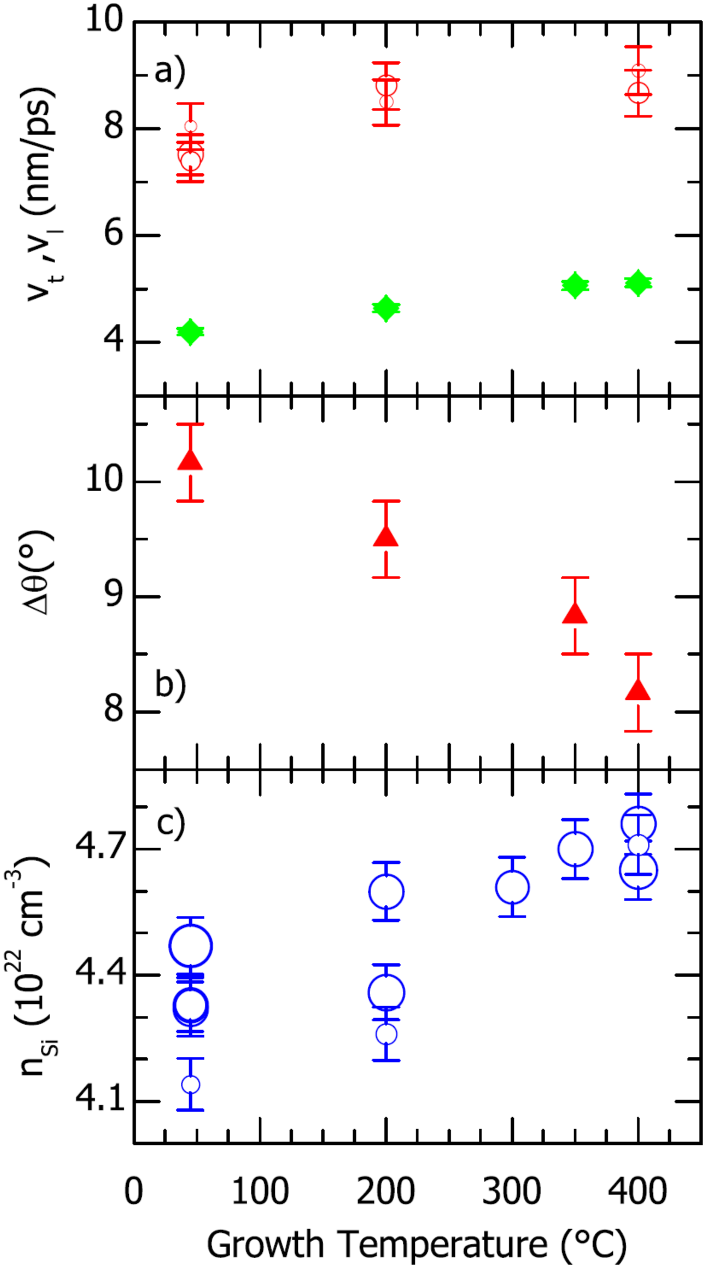
Experimental Observation for a-Si

- Internal friction of films deposited at high temperatures T_s
  - *very* different from film annealed at same temperature
  - $\phi \sim 2\text{x}10^{-6}$ vs $\sim 10^{-4}$
- Improves steeply around $T_s \sim 400$ C



X. Liu, F. Hellman, et al, *PRL* **113**, 025503 (2014)

Other Properties of a-Si Modified for High T_s



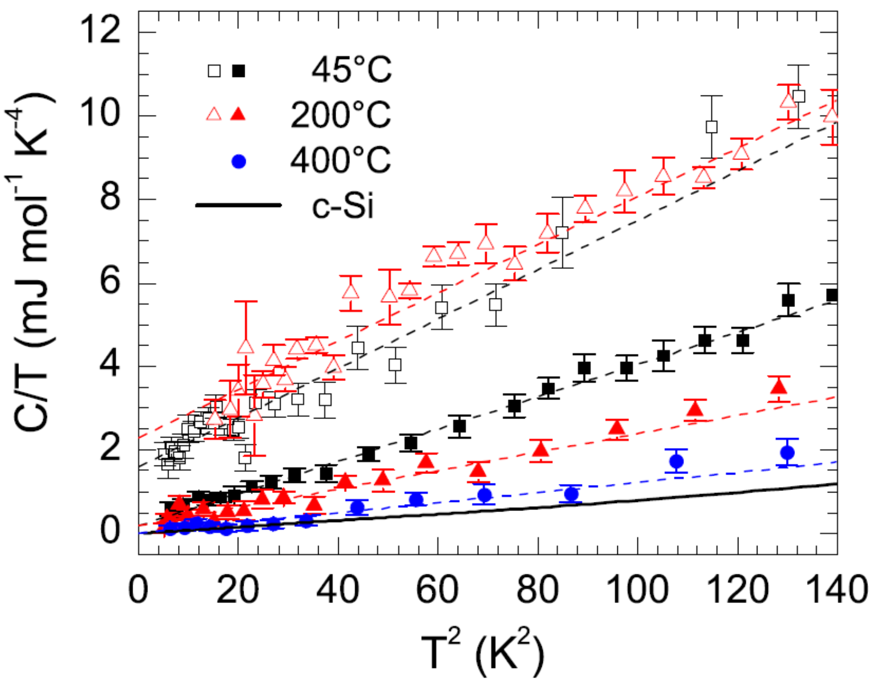
sound velocity approaches asymptote
(via paddle resonance)

distribution of bond angles narrows
(via Raman lineshape)

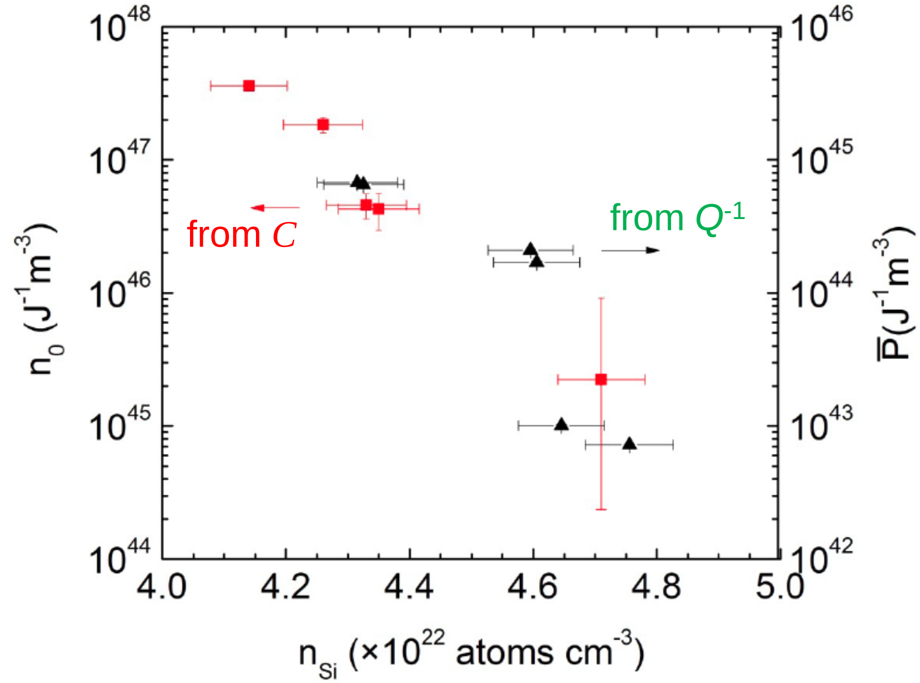
density increasing
(via RBS)

D.R. Queen, F. Hellman, et al,
Journal of Non-Crystalline Solids **426** 19–24 (2015)

Heat Capacity Approaches Crystalline Silicon Value



same n_{TLS} from Q^{-1} and C



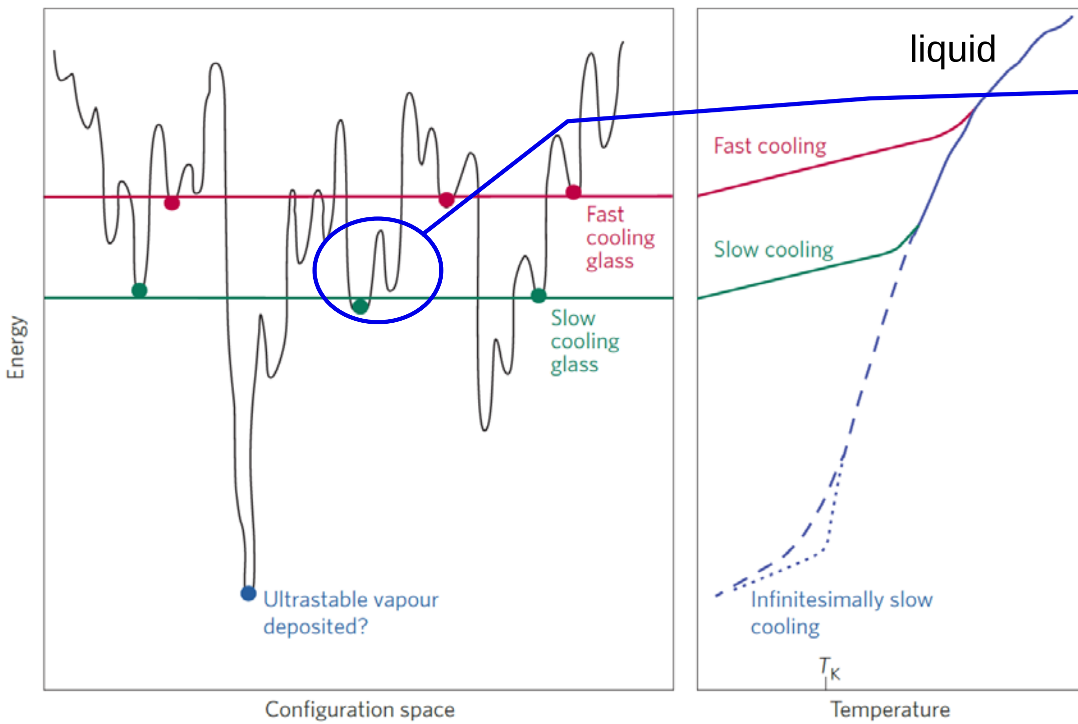
both correlate with film density

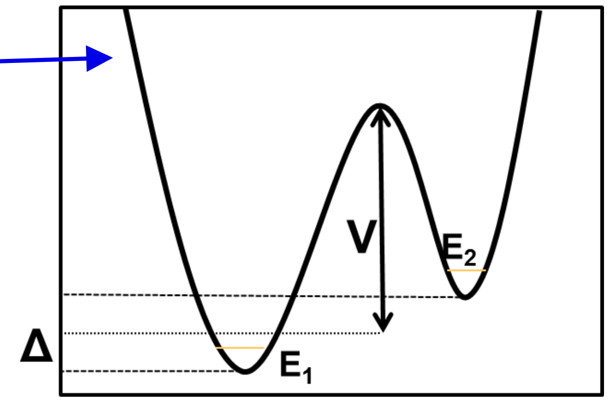
Material properties suggest greatly reduced n_{TLS} for high T_s

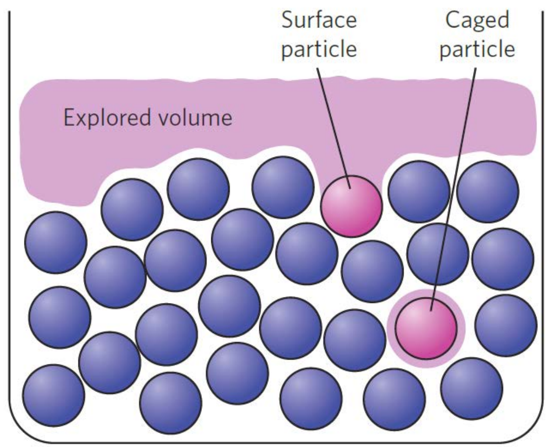
What is the mechanism?

D.R. Queen, F. Hellman, et al,
Journal of Non-Crystalline Solids **426** 19–24 (2015)

Energy Landscape of Amorphous Materials







- Slower cooling liquid reaches lower energy states
  - lowest energy states inaccessible from liquid
- Vapor phase deposition accesses lower energies
  - simulations* suggest surface liquid layer has orders of magnitude higher mobility than caged particles in solid
  - deposition at high temperature more effective than post-annealing

figures from Parisi, *Nature Mater.* **12,** 94 (2013)

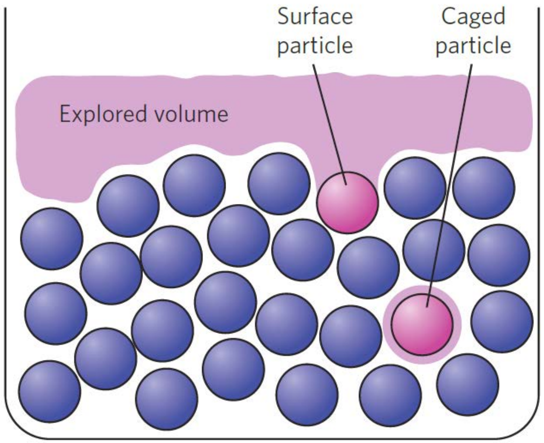
* S. Singh, *Nature Mater.* **12,** 139 (2013)

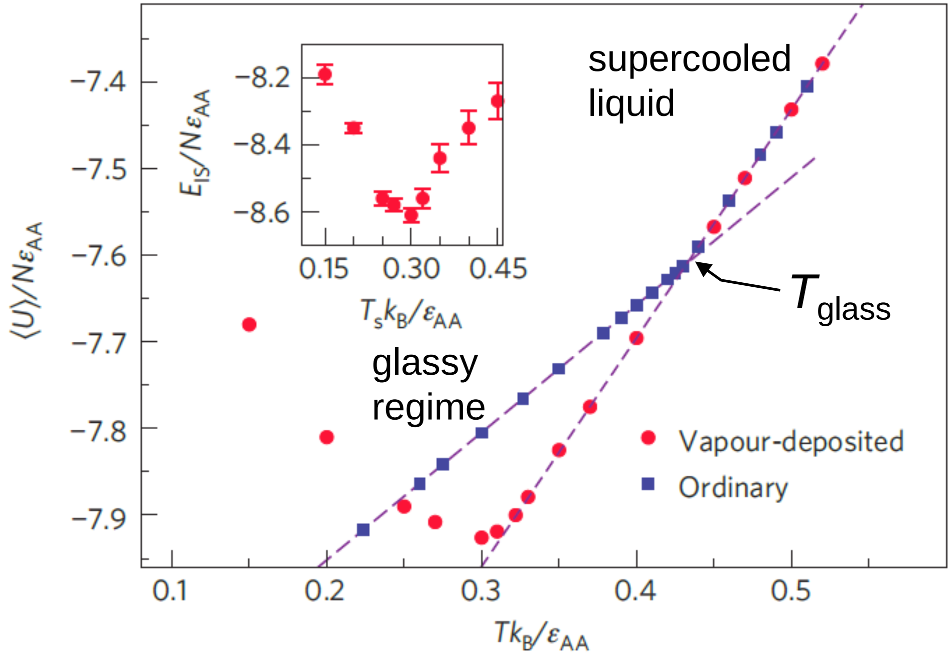
Simulation of Model System

S. Singh, *Nature Mater.* **12,** 139 (2013)

- Add (numerically) Lennard-Jones particles
  - one-by-one onto free surface
  - ~5000 particles
  - explore effect of substrate temperature
  - compare to MD simulation of cooling liquid

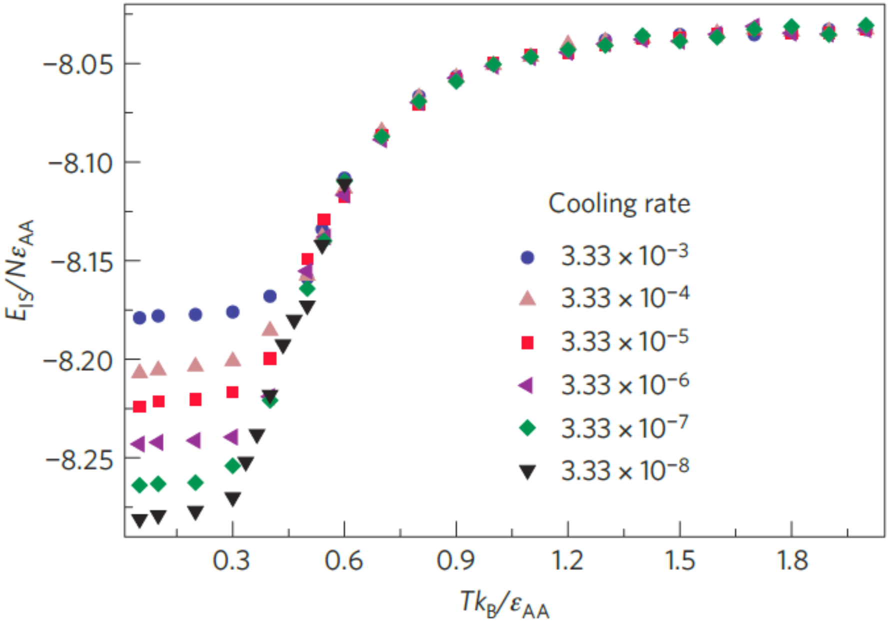
liquid vs vapor deposition)





reach more stable glass from vapor

Structure energy (from cooled liquid)



slow cooling $\Rightarrow$ more stable

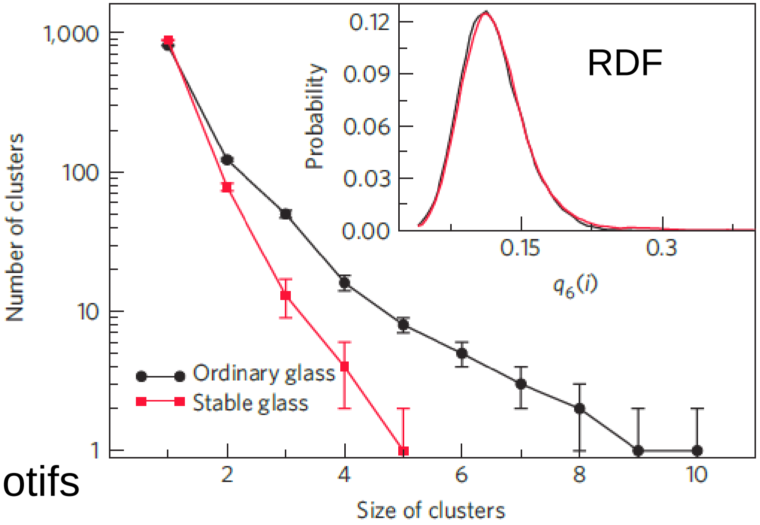
"Ordinary" vs "Stable" Glass

- "Ordinary" glass: obtained by cooling liquid
- "Stable" glass: vapor deposited at T_s yielding lower energy configuration than ordinary glass

- Lowest energy obtained for $T_s \sim 0.8\ T_{glass}$
  - unattainable from liquid at lab time scales
  - presumably also not from annealing film vapor deposited at a lower T_s

- Densities 1-2% higher than ordinary glass

- Structural features
  - radial distribution functions indistinguishable
  - more clustering of "ordered particles" in ordinary vs stable glass
  - stable glass more homogeneous ordinary glass has wider range of packing motifs



Compare Model Calculations to Expt.

- a-Si expt.: steep improvement for $T_s \sim$ 400 C
  - theoretical* $T_{\mathrm{glass}} \sim 900$ K :

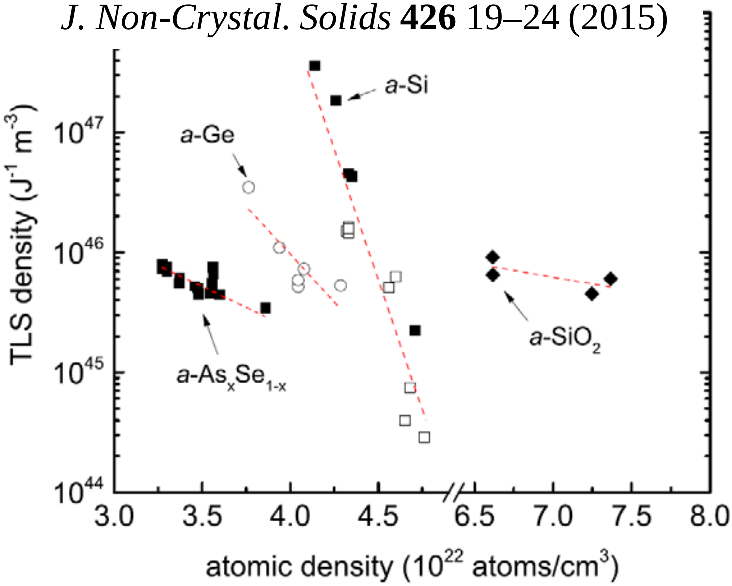
    critical $T_s / T_{\mathrm{glass}} \sim 0.75$ vs predicted $T_s \sim 0.8\ T_{glass}$
  - similar trends seen in some organic glasses

    T. Perez-Castaneda, *PNAS* **111**, 11275 (2014)

* C.R. Miranda and A. Antonelli, *J. Chem. Phys.* **120**, 11672 (2004)

- Low TLS correlates with high density

D.R. Queen, F. Hellman, et al, *J. Non-Crystal. Solids* **426** 19–24 (2015)



- Rayleigh scatter in SiO_2
  - decreases with anneal time

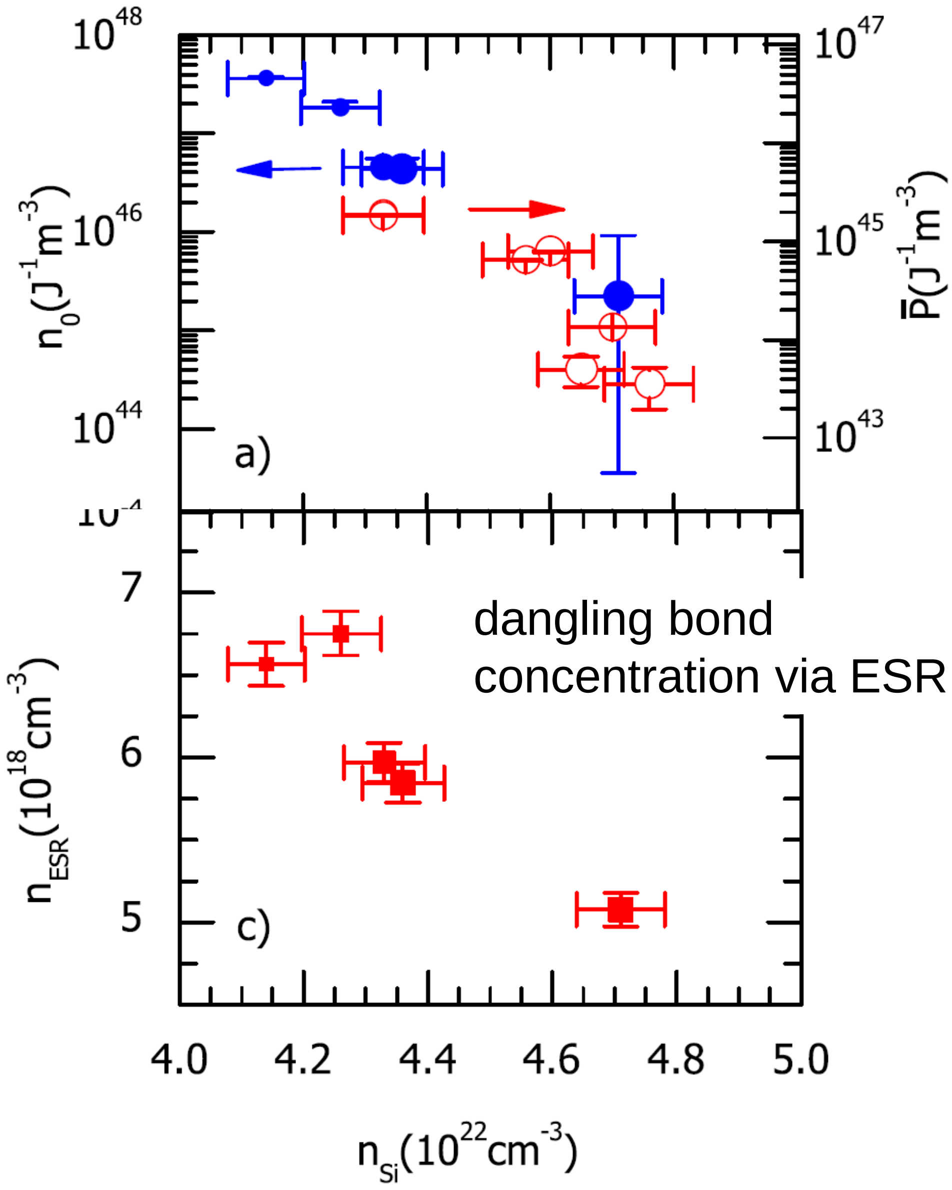
    and anneal temperature

A. Ageev, S. Penn, P. Saulson, *Class. Quantum Grav.* **21** (2004) 3887–3892

- Hellman posits evidence for heterogeneous structure:
  - dense backbone network surrounding low density regions
  - TLS and dangling bonds in the low density regions

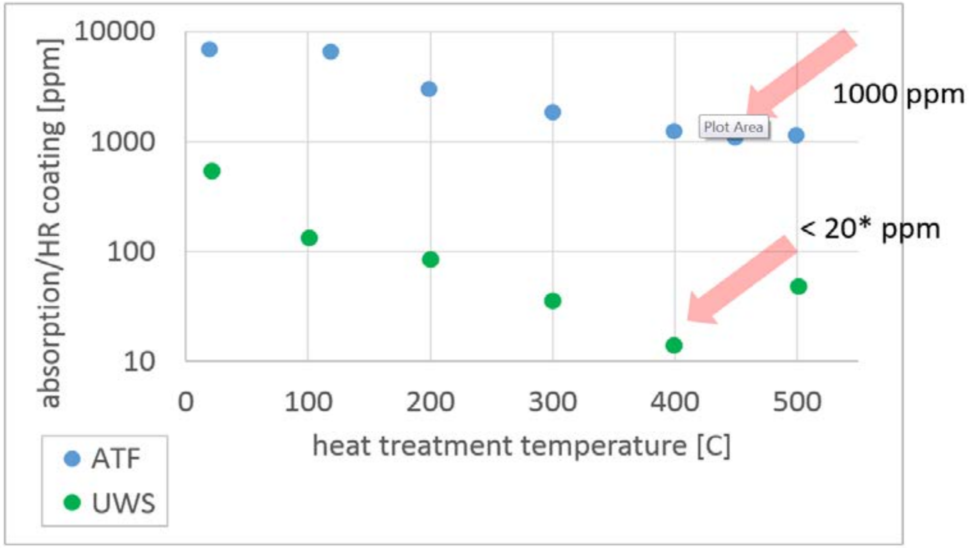
Optical Absorption in a-Si a Remaining Issue

- Absorption from dangling bonds too high
- Dangling bonds also reduced by high T_s
  - but ~2x, not 10^3x



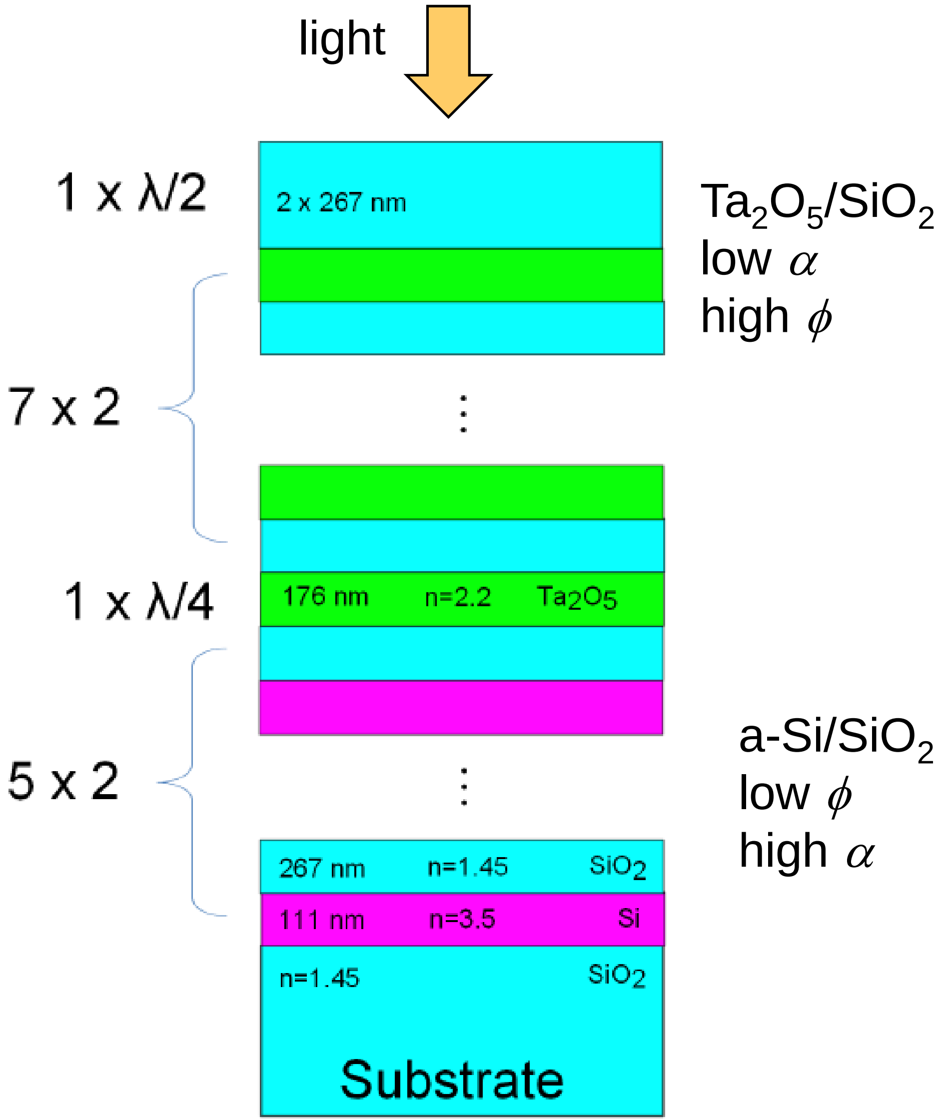
Optical Absorption in a-Si a Remaining Issue

- Absorption from dangling bonds too high
- Dangling bonds also reduced by high T_s
  - but ~2x, not 10^3x
- Recent IBS promising
  - alternative method to increase surface mobility
  - Stuart Reid 10:00 Wed.



Optical Absorption in a-Si a Remaining Issue

- Absorption from dangling bonds too high
- Dangling bonds also reduced by high T_s
  - but ~2x, not 10^3x
- Recent IBS promising
  - Stuart Reid 10:00 Wed.
- Multi-material coatings could mitigate absorption
  - Steinlechner poster



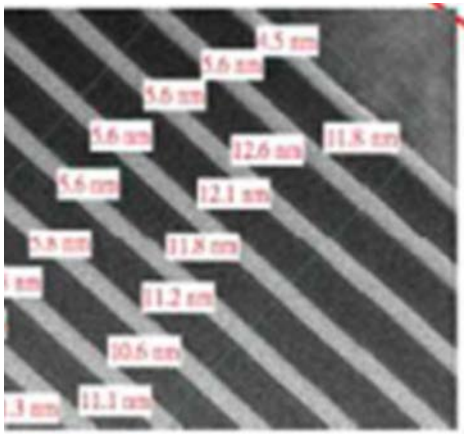
J. Steinlechner, et al , *Phys. Rev. D* **91** 042001 (2015)
W. Yam et al, *Phys. Rev. D* **91**, 042002 (2015)

Towards a Better Glass

- "Stable glass" concept
  - appears to apply for a-Si and organic glasses

- Extendable to amorphous oxides?
  - no data yet for high temperature deposition
    - experiments in preparation
      - high glass temperatures potential challenge
  - Ion-beam sputter/assist
    - (controversial?) proposed alternative route to high surface mobility

- Alternative: higher annealing temperatures for oxide glasses
  - push toward more uniform glass
  - suppress devitrification
    - suitable stabilizing dopant, e.g. Zr:Ta_2O_5
    - nanolayers: poster A. Amato
      - suppress crystallization
      - interface scatter an issue?



Atomic Structure

- Characterization of structure of amorphous materials more difficult than crystalline
  - several applicable techniques:

- Short-range order (<1 nm):

- TEM (transmission electron microscopy)
  - Data: Nearest atomic neighbor distribution
  - Pro: Can be used on any amorphous coating, multi or single layer with layer dependent data
  - Con: Requires destructive sample prep

- XAFS (X-ray absorption fine structure)
  - Data: Atomic species specific structure (around only Ta, Ti)
  - Pro: No sample prep
  - Con: Requires synchrotron with limited availability (3 X per year)

- NMR (nuclear magnetic resonance)
  - Data: Local atomic co-ordination (predominantly O)
  - Pro: Provides direct measurement of coordination number
  - Con: Required destructive sample prep requires coatings on salt substrates

- Raman spectroscopy
  - Data: Bond angle changes, Chemical ID, Phonon spectra
  - Pro: Probes vibrational modes vs static regime of other techniques
  - Con: Requires models to fully interpret

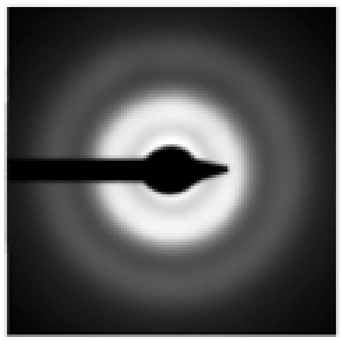
Atomic Structure

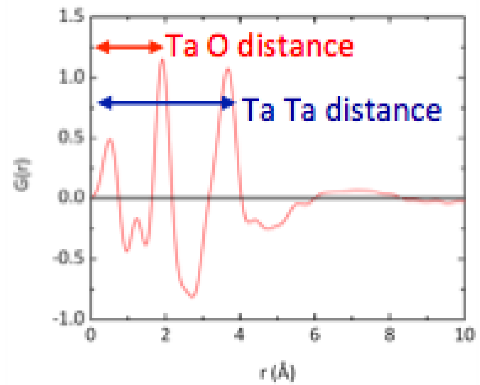
- Medium-range order (0.5 – 5 nm):
  - FEM (fluctuation electron microscopy)
    - Data: Quantifies the MRO by measuring variance in TEM diffraction patterns
    - Pro: MRO can be measured at various probe sizes (MRO at each set electron beam size)
    - Con: Destructive TEM sample prep, difficult to model
  - GI-XPDF (grazing incidence X-ray pair distribution function)
    - Data: Nearest neighbor distribution with resolution into medium range order
    - Pro: No sample prep required, depth dependence
    - Con: Requires synchrotron time

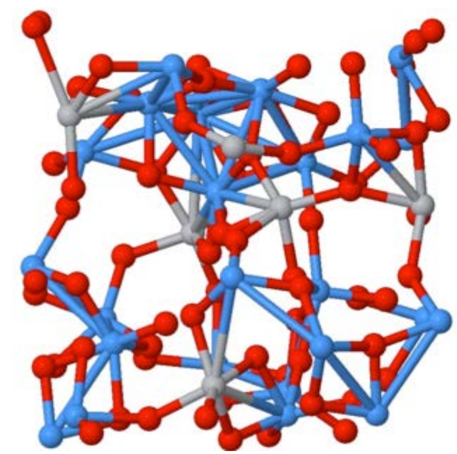
- talk by Riccardo Bassiri 9:40 Tuesday

Example of Atomic Structure Measurement

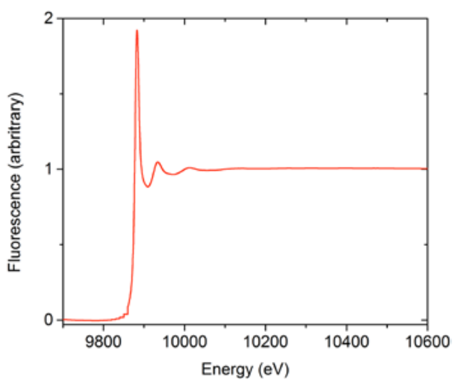
- Pair Distribution Function (PDF), G(r), electron or X-ray diffraction

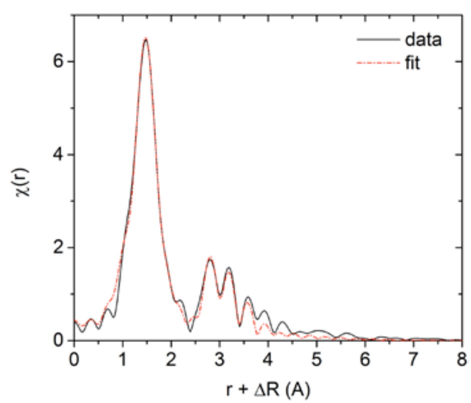






- Extended X-ray Absorption Fine Structure (EXAFS),
  - Ta L_{III} edge





How to Use Structural Characterization

- Some observations:
  1. structural changes with annealing and doping are subtle
  2. difficult to unambiguously invert from data to structure

- Some useful topics for discussion:
  - given progress in theoretical modeling, how to leverage data from structural probes?
  - forward vs inverse calculations for structural data

Some Current Directions in Coating Research

- Ultra-stable oxide glasses: experiments beginning
  - high temperature deposition
  - ion beam sputtering/ion assisted
  - slow deposition rates

- Other experimental approaches to synthesize lower loss coatings:
  - nanolayer stabilization
  - dopant-stabilized high-temperature annealing
  - multi-material coatings

- Atomic structure modeling/characterization
  - experiment and theory reaching maturity to usefully interact
  - tie-up loss and structure modeling with measurements
  - explore theoretically advantageous material/dopant combinations

Some Other Topics for Discussion

- Challenges
  - 4? year timeline to better coatings for A+

    clarify performance goals and timeline
  - need to maintain long-range (cryo) research while targeting near-term (RT)
  - materials discovery not easily schedulable
  - need time for coating vendor to implement proposed coating once discovered
- How to tie up synthesis, loss measurements, structural models, coating design, and expts
  - more effective coordination across collaboration
  - complex decision space

    how to reach decision points on what not to do?