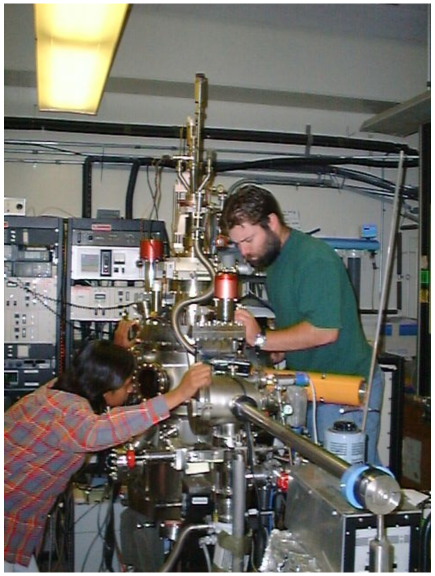
Two level systems and thermal noise

why do coatings have this noise and how do we reduce it

Frances Hellman

University of California, Berkeley

New LIGO Coatings group member



UHV growth chambers for prototyping materials

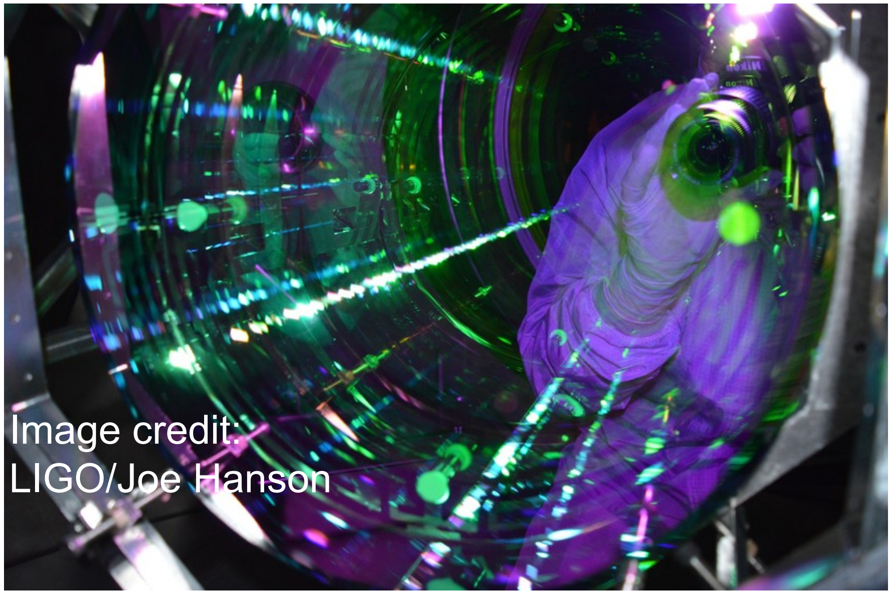
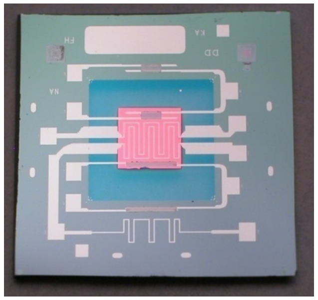


Image credit: LIGO/Joe Hanson



Nanocalorimeters: Si micromachined chips for thin film heat capacity

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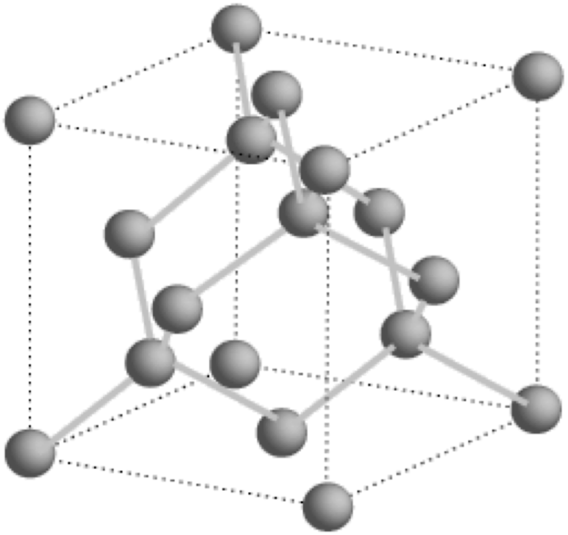
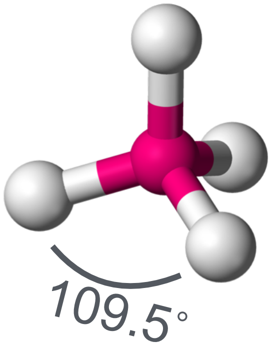
Current mirror coatings are amorphous (=non-crystalline)
No long range structural order, but have short range order

Traditionally, glasses quenched from liquid
Also can be made by vapor deposition
Structure of Silicon



Xtal Si: diamond structure



Amorphous Si: still has tetrahedral coordination

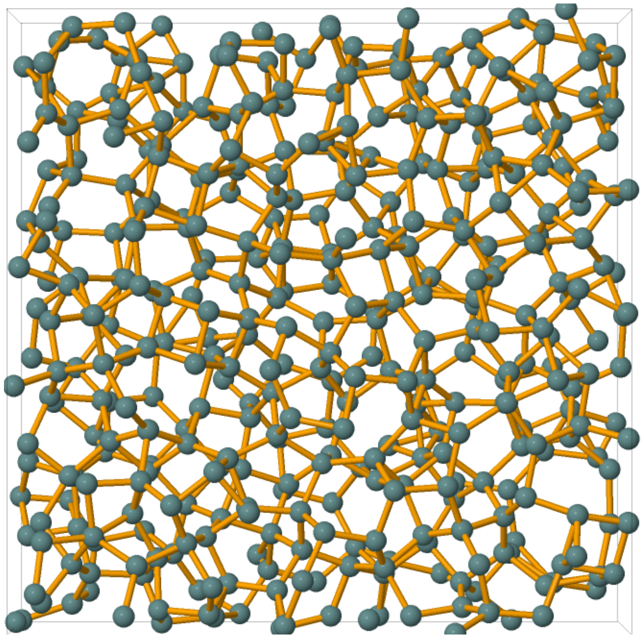
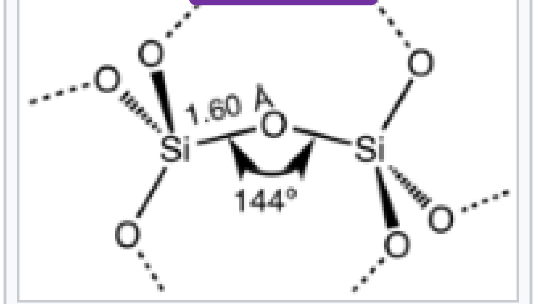


Image credit: Kiran Prasai

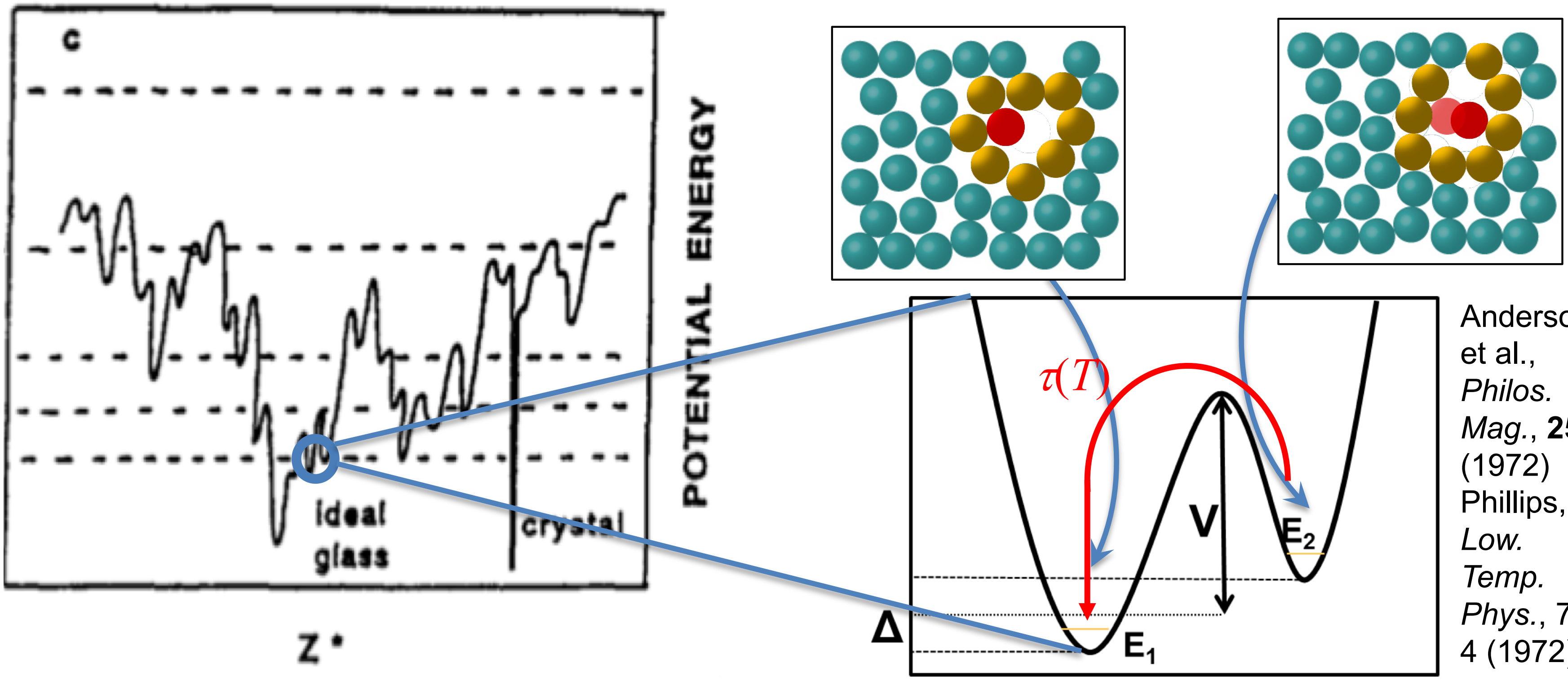
SiO_2



O-Si-O bonds are fixed angle but Si-O-Si angle is quite floppy – different energy scales

Energy landscape of configurations: "nearby" minima lead to tunneling *or* thermally-activated motion of groups of atoms

C.A. Angell, Physica D 107, 122 (1997)



Anderson, et al., *Philos. Mag.*, **25** 1 (1972)
Phillips, *J. Low. Temp. Phys.*, 7 3-4 (1972)

Two-Level Systems from neighboring energy minima in structural landscape:

- At low T, atomic structure **tunnels** between these ➢ μeV energy splitting $E_{1,2} \pm \Delta$
- At higher T, atomic motion is **thermally activated**, requiring $k_B T$ ~ barrier height V

In both cases, atomic motion leads to dissipation (thermal noise)

For a single V, dissipation at frequency ω will have a peak at temperature T

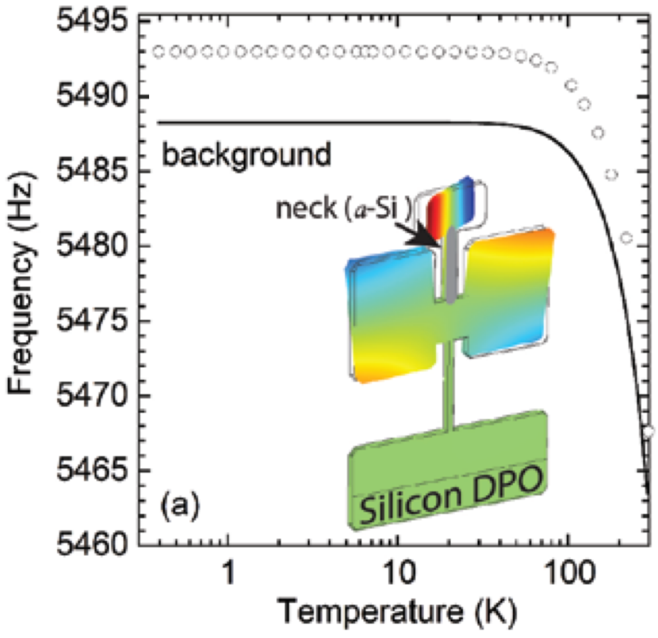
➢ at 1 kHz, 0.5 eV barrier heights has a peak ~ room temperature

50 meV barrier heights has a peak ~ 30K

A *distribution* of μeV tunneling-induced energy splitting leads to T-independent losses

Internal friction to measure losses

Measure resonant frequency and internal friction (damping) Q^{-1} of a xtal Si double paddle oscillator (DPO) or cantilever as a function of T before and after depositing a film, or a silica disk at room temperature using GeNS (gentle nodal suspension)



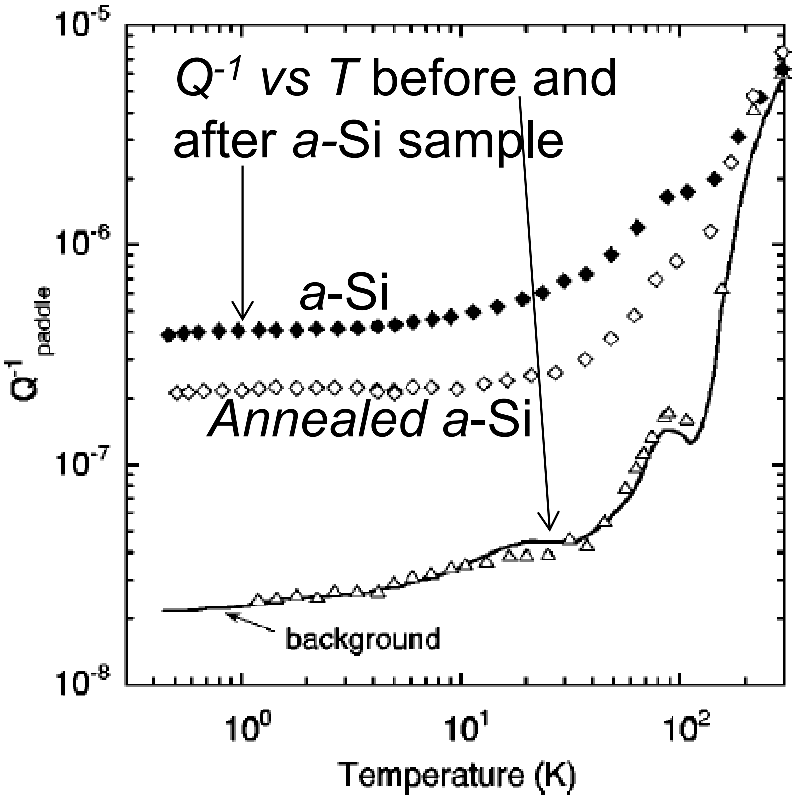
X. Liu and R.O. Pohl, Phys. Rev. B **58**, 9067 (1998)

Internal friction $Q^{-1}(T)$ has a low T plateau due to *tunneling two level system* (TLS)-phonon interactions

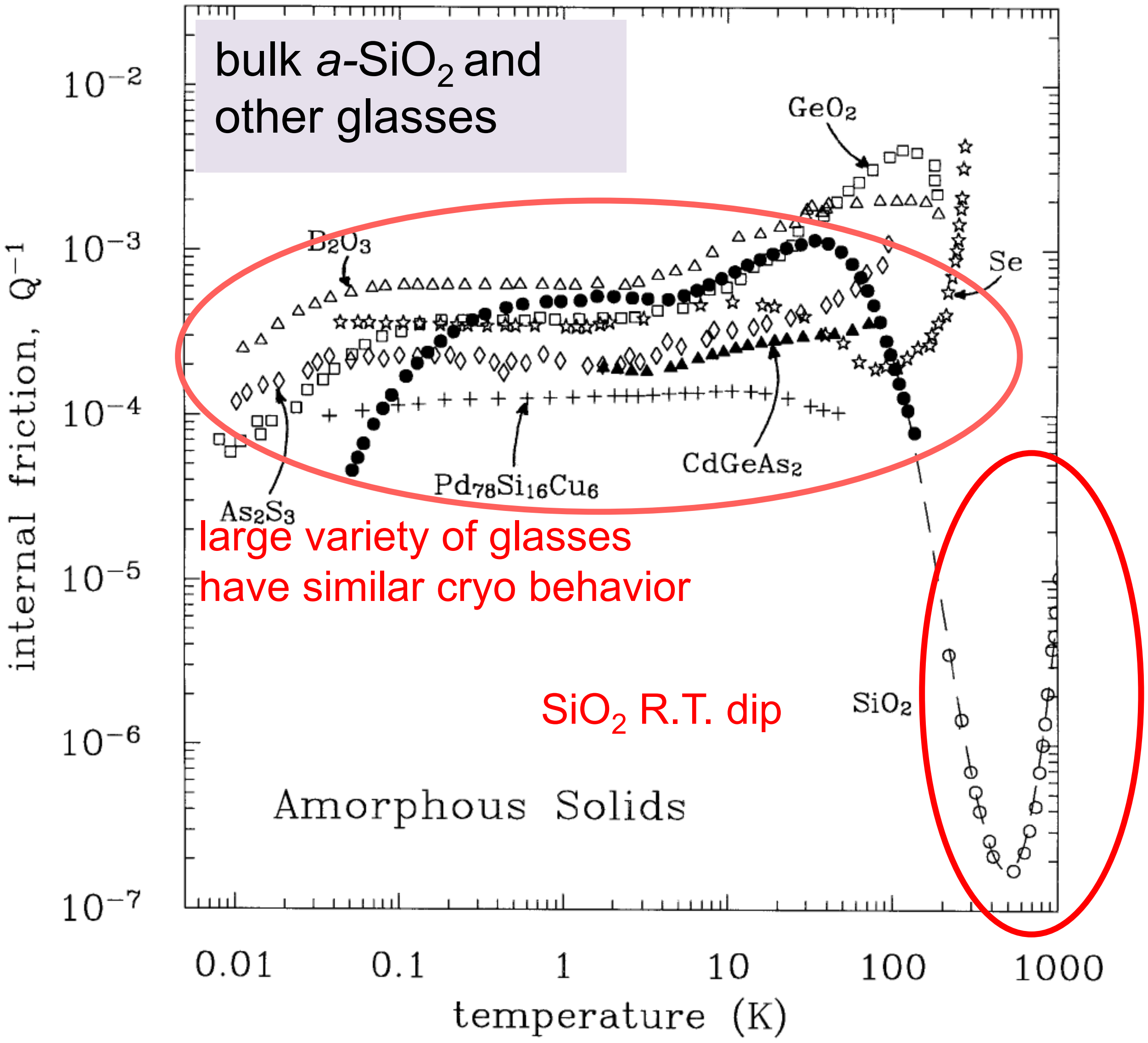
Q_o^{-1} proportional to $\bar{P}$ (density of TLS) with a ***poorly understoood TLS – phonon coupling parameter*** γ, also called the deformation potential

$$Q_0^{-1} = \pi \bar{P} \gamma^2 / 2\varrho v^2$$

ρ is density, and v is sound velocity

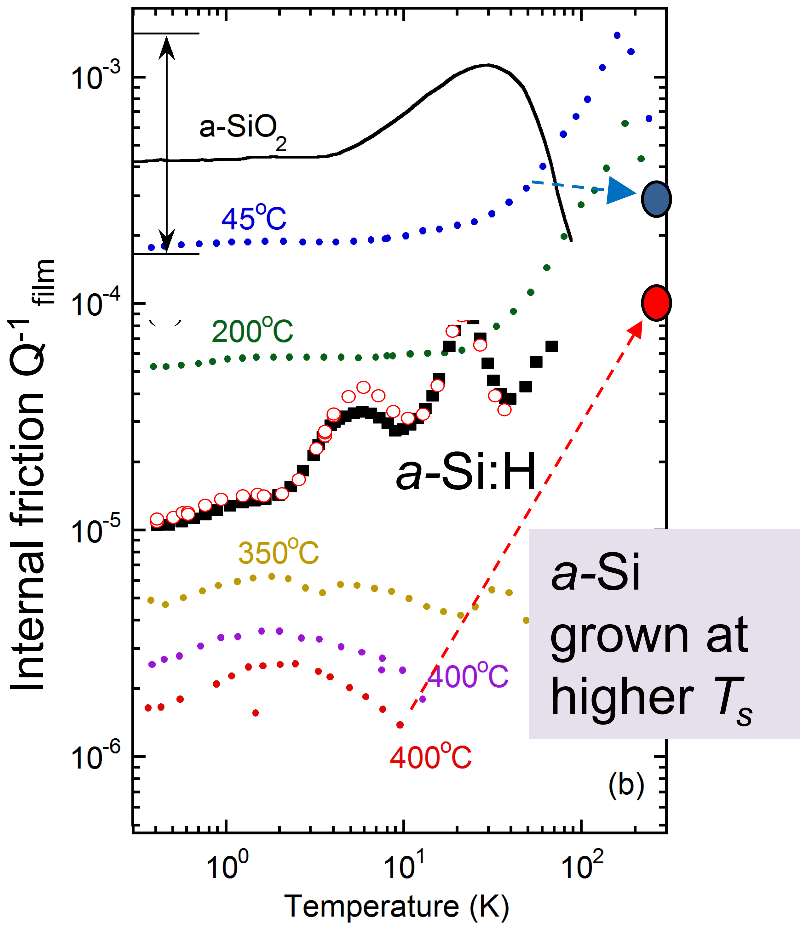

Q^{-1} *vs T* before and after *a*-Si sample

"Universal" mechanical losses at low T (tunneling)
Higher T more variable, including peaks (thermally activated)



K.A. Topp, *Z. Physik B Condensed Matter* 101 235–45 (1996)

Thin film amorphous Silicon: internal friction Q^{-1} (and excess heat capacity) is strongly reduced (decades) by increased growth T_s

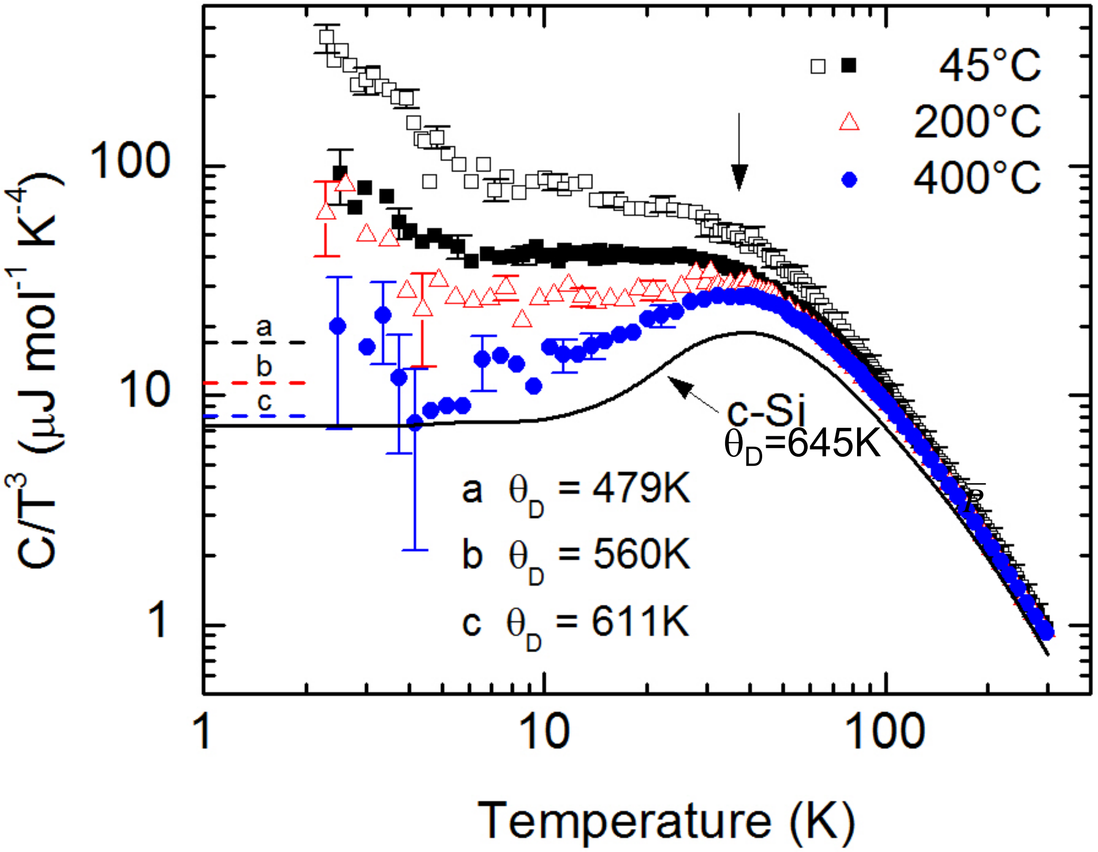


a-Si grown at higher T_s

- *a*-Si films grown at lower T_s have typical (high) Q^{-1} (like *a*-SiO_2)
- ***a*-Si films grown at 400˚ C have *very low Q^{-1} at low T (perhaps good for LIGO Voyager); (at RT better but only 4x)***
- BUT, large absorption – likely need H to eliminate dangling bonds in *a*-Si
- *a*-Si:H has low Q^{-1} (although peaks not well understood) BUT *has high excess low T heat capacity indicating local structures that cause loss are present, just not coupling to acoustic waves – why and what does this do to higher temp loss???*

D. R. Queen, X. Liu, J. Karel, T.H. Metcalf, F. Hellman, Phys. Rev. Lett. 110, 135901 (2013).
X. Liu, D. R. Queen, T. H. Metcalf, J. E. Karel, F. Hellman, Phys. Rev. Lett. 113, 025503 (2014).
D.R. Queen, X. Liu, J. Karel, Q. Wang, R.S. Crandall, T.H. Metcalf, F. Hellman, Eur. Phys. Lett. **112**, 26001 (2015).
M. Molina-Ruiz, H. C. Jacks, D.R. Queen, T. Metcalf, X. Liu, Q. Wang, R. S. Crandall, F. Hellman, in preparation

Variable TLS in *a*-Si also in specific heat *C*: excess *C* at low T



Linear term in C: $c_1 \sim n_0$
Excess T^3 *term* (c_{ex})

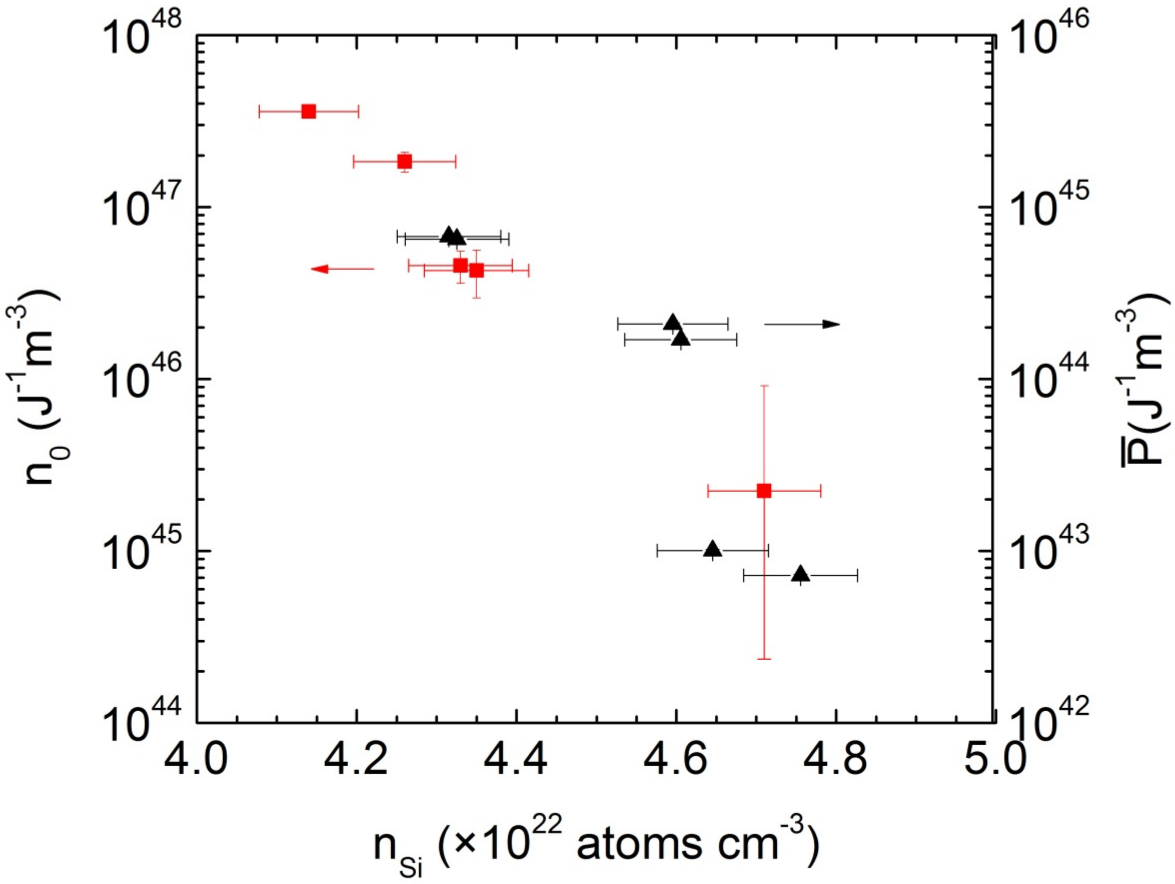
Films grown at 400° C have *C(T)* only a little above *c*-Si; small n_0 and small c_{ex} (Also, thermal conductivity shows no plateau)

Films grown at lower T_s have excess Q^{-1} and *C(T)* above Debye value (from transverse and longitudinal sound velocity measurements)

Fit low T *C(T)* to $c_1T + C_3T^3$; both n_0 and c_{ex} depend on T_s *and on film thickness*

Sound velocity, bond angle distribution, nanovoid size depend on T_s *&* ***not*** *thickness*

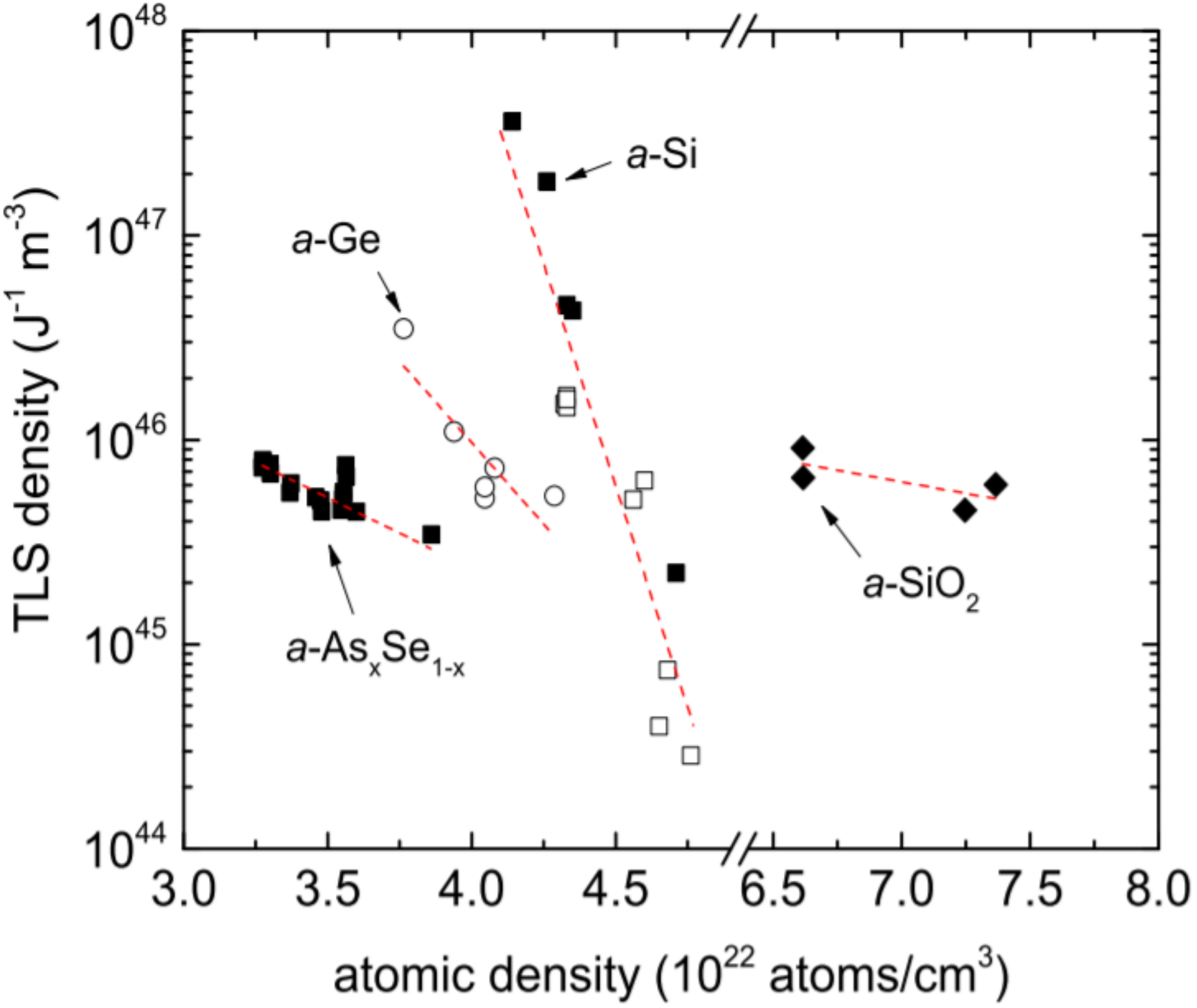
TLS density from specific heat and internal friction are proportional to each other, *and depend on film density*



Crystalline Silicon:
$n_{Si}=5\times10^{22}$ cm^{-3}

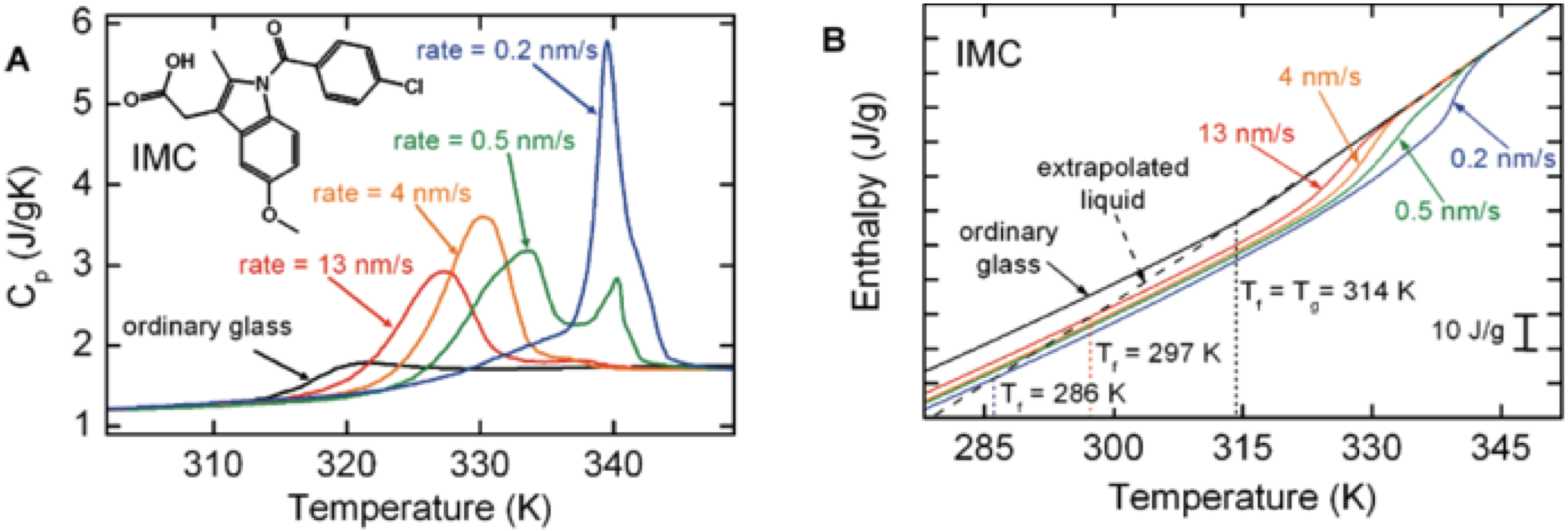
- n_o *and* $\overline{P}$ *vanish as* $n_{Si} \rightarrow n_{crystalline\ Si}$
- $n_0/\overline{P}$ ~ 8 – similar to other glasses. n_o and $\overline{P}$ proportional in usual TLS model
- TLS vanish with increasing n_{Si} – associated with low density regions/nanovoids??
- Correlation is over nearly 3 decades

TLS (either n_o or $\bar{P}$) dependence on atomic density seen in a range of amorphous materials



From D.R. Queen et al., JNCS 426 (2015), 19-24

Vapor deposited films of indomethacin (IMC); ultrastable glasses *with low TLS grown at T_s < glass transition T_g*



Heat capacities and enthalpies for vapor deposited glasses of indomethacin (IMC) with decreasing deposition rates; grown at "magic" $T_s \sim 0.8\ T_g$. As rates are lowered, T_f decreases, as does enthalpy, indicating a more stable glass. Similar effects seen with growth T.

These films also have low TLS!!

T. Perez-Castaneda, C. Rodriguez-Tinoco, J. Rodriguez-Viejo, M.A. Ramos, "Suppression of tunneling two-level systems in ultrastable glasses of indomethacin," PNAS 111(31), 11275 (2014)

M.D. Ediger, "Vapor-deposited glasses provide clearer view of two-level systems," PNAS 111(31), 11232 (2014).

Hypotheses re vapor deposited *a*-Si

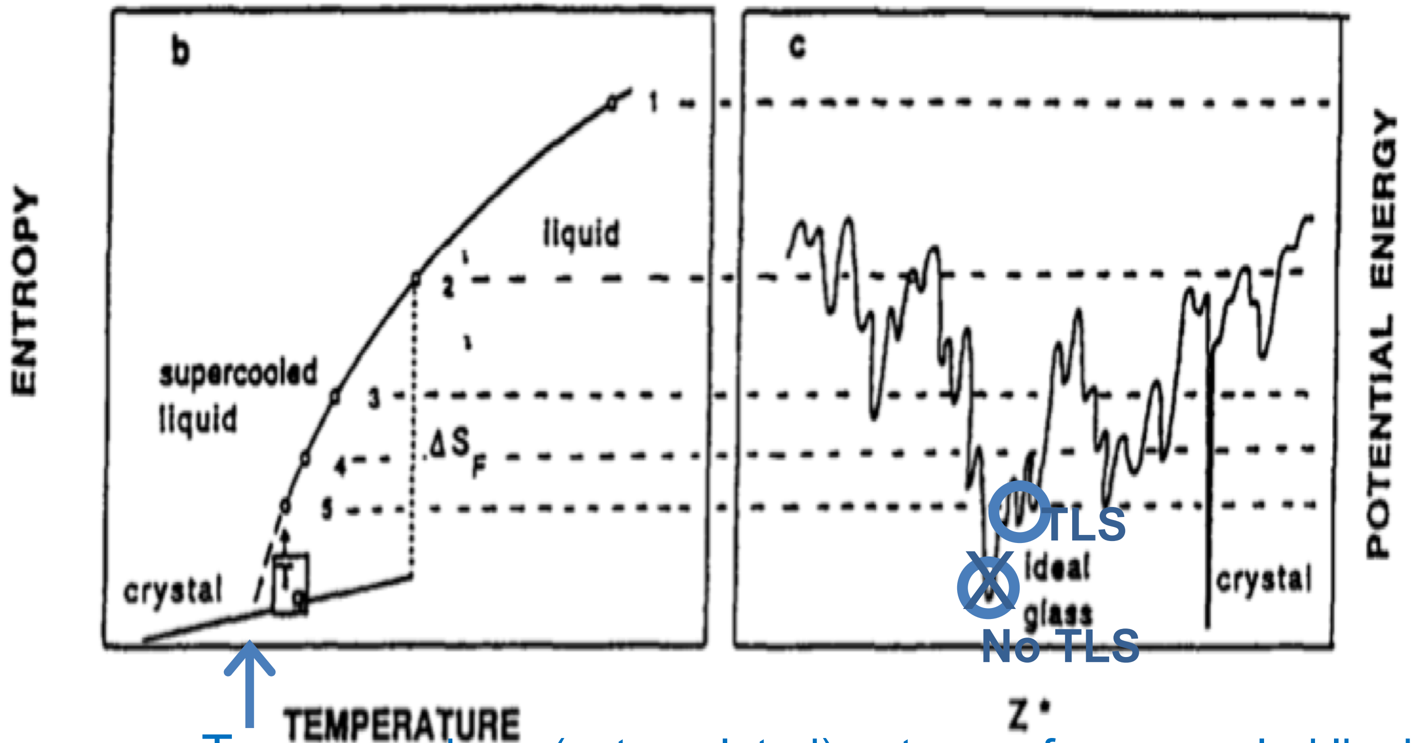
Vapor deposited films of covalent materials such as *a*-Si or *a*-SiO_x (or Alumina or SiNx or Tantala) have to date not been probed for ideality/ultrastability

The glass transition of a-Si has never been measured (because it can't be quenched) but theory suggests 850K (C.R. Miranda and A. Antonelli, J. Chem Phys 120, 11672 (2004).

Our growth T to get low TLS is 673K ~ 0.8 T_g!! (similar to IMC work)

We have also seen effects of deposition rate and thickness on density and TLS, similar to IMC work; TLS measurements in progress on these other films

Connection between energy landscape, entropy, and TLS



C.A. Angell, Physica D 107, 122 (1997)

$T_{Kauzmann}$ where (extrapolated) entropy of supercooled liquid crosses crystal!

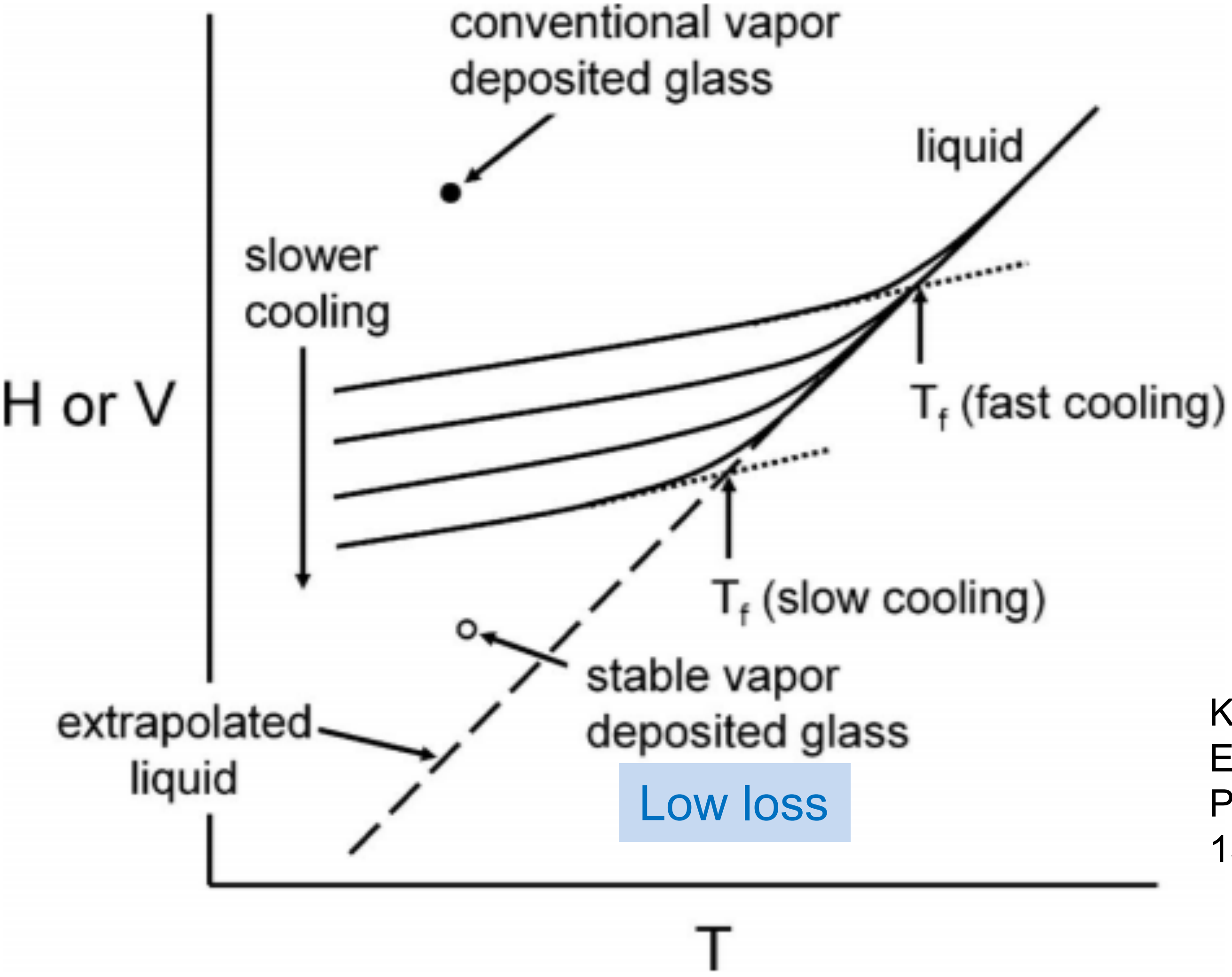
The energy landscape (right) as related to the glass transition of a liquid (left). Glasses falling out of the equilibrium supercooled liquid at a given dashed line correspond to configurations in the energy landscape.

Hypothesis: vapor deposition offers a way to directly access low lying (ideal) glass state
Due to high atomic mobility at film growth surface despite being at low T.

Hypothesis: Ideal glass has no nearby energy minima, so no TLS, unlike most other states

Enthalpy or Volume (density) as a function of T starting from liquid Vapor deposited compared to liquid quenching of amorphous material

High loss



Low loss

H and V based on simulations of atoms with simple bonds (Lennard-Jones). Kiran Prasan will talk about *a*-Si.

Kearns, Swallow, Ediger, J. Chem. Phys. 127, 154702 (2007)

Could this apply to vapor deposited amorphous SiO_2? Does this lower losses at all T, some T, for all materials?

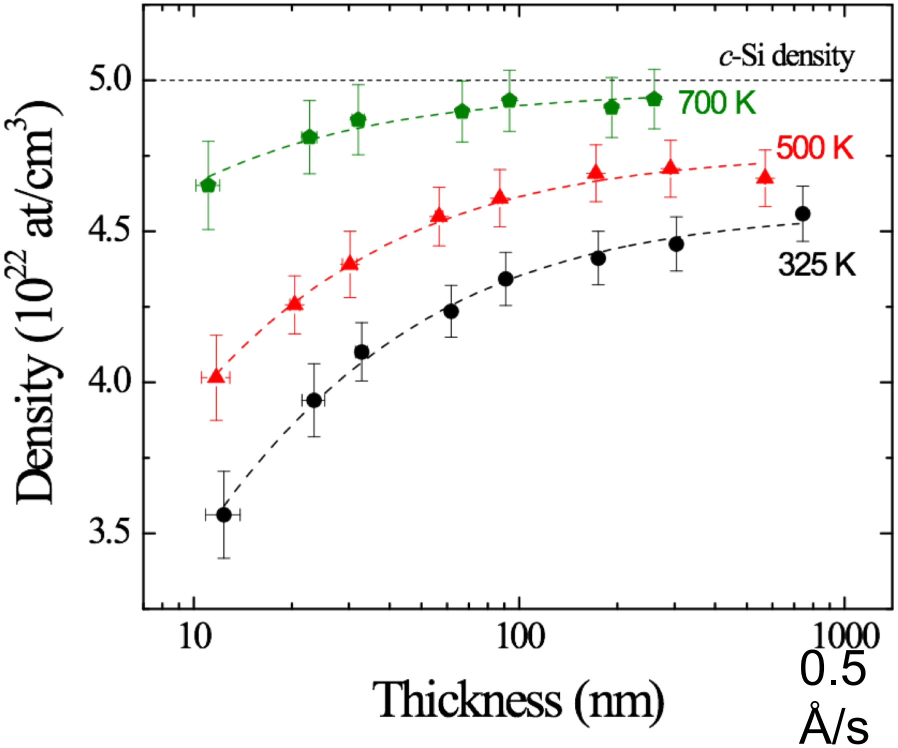
Comments and Open questions

- Random first order transition (RFOT) theory of glass transition predicts divergence of correlation length ξ of supercooled liquid as $T \rightarrow T_K$
- Also predicts TLS density ~ ξ^{-3} , supporting the hypothesis that TLS density is highly suppressed for deposition T near T_K
- But, surface diffusion needs to be fast near T_K , argues T_K needs to be ~T_g, (which means "fragile" glasses like *a*-Si and indomethacin, not *a*-SiO_2)
- *But, a*-SiO_2 has a high density of low energy floppy modes, so perhaps

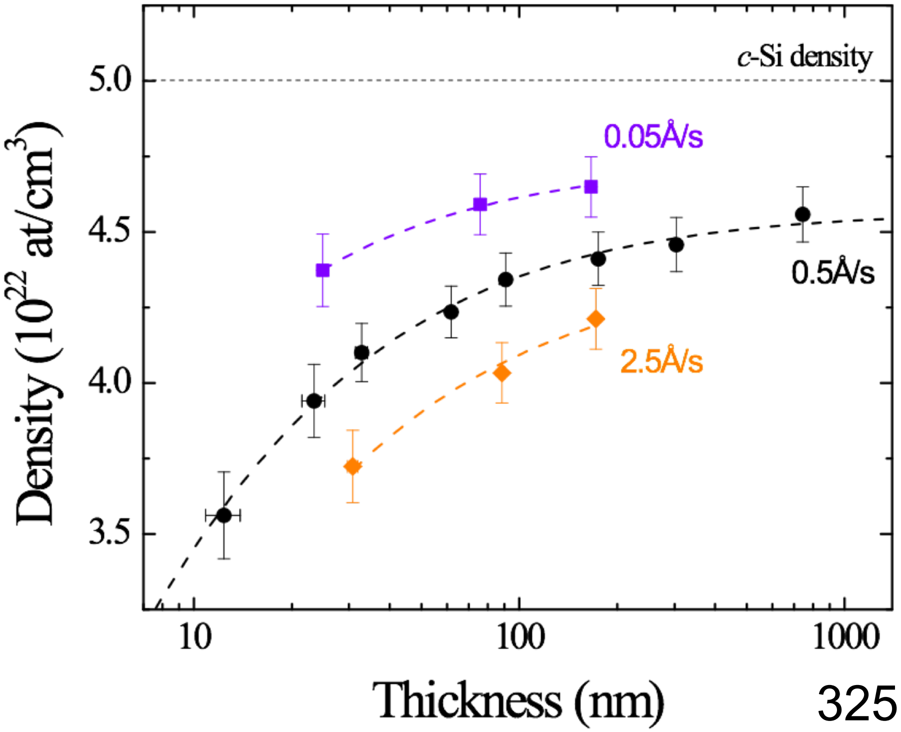
OPEN QUESTIONS

- Is low TLS related to growth near T_K? (If (and only if) surface mobility during growth is high). Fragile glasses have T_K near T_g, where mobility is high, so low TLS would be correlated with fragility
- Or is low TLS related to nature of bonding: overconstrained (tetrahedral Si) versus underconstrained (Si-O-Si bonds in SiO_2)
- Test with amorphous SiO_2, Ta_2O_5, Al_2O_3, and Se_xGe_{1-x} alloys!!
- What is the relationship to density in the IBS overdense films, which *could* be higher in the energy landscape with higher TLS.

Growth parameters substantially modify amorphous Si film density and some measures of structure



0.5 Å/s



325K

- Thickness, growth temperature T_g, and growth rate affect film density and roughness; room T growth flattest for all thicknesses; higher T_g thin is flat, roughens with thickness (1.5 nm RMS at 300 nm)
- Thinner, low growth T, high growth rate films are less dense
- On what length scale(s) do density changes occur? Little variation in dangling bond density or macroscale structure
- *Variations in bond angle disorder*, *medium range order, nanovoid size and number* (Raman, Fluctuation Electron Microscopy, positron doppler broadening spectroscopy)

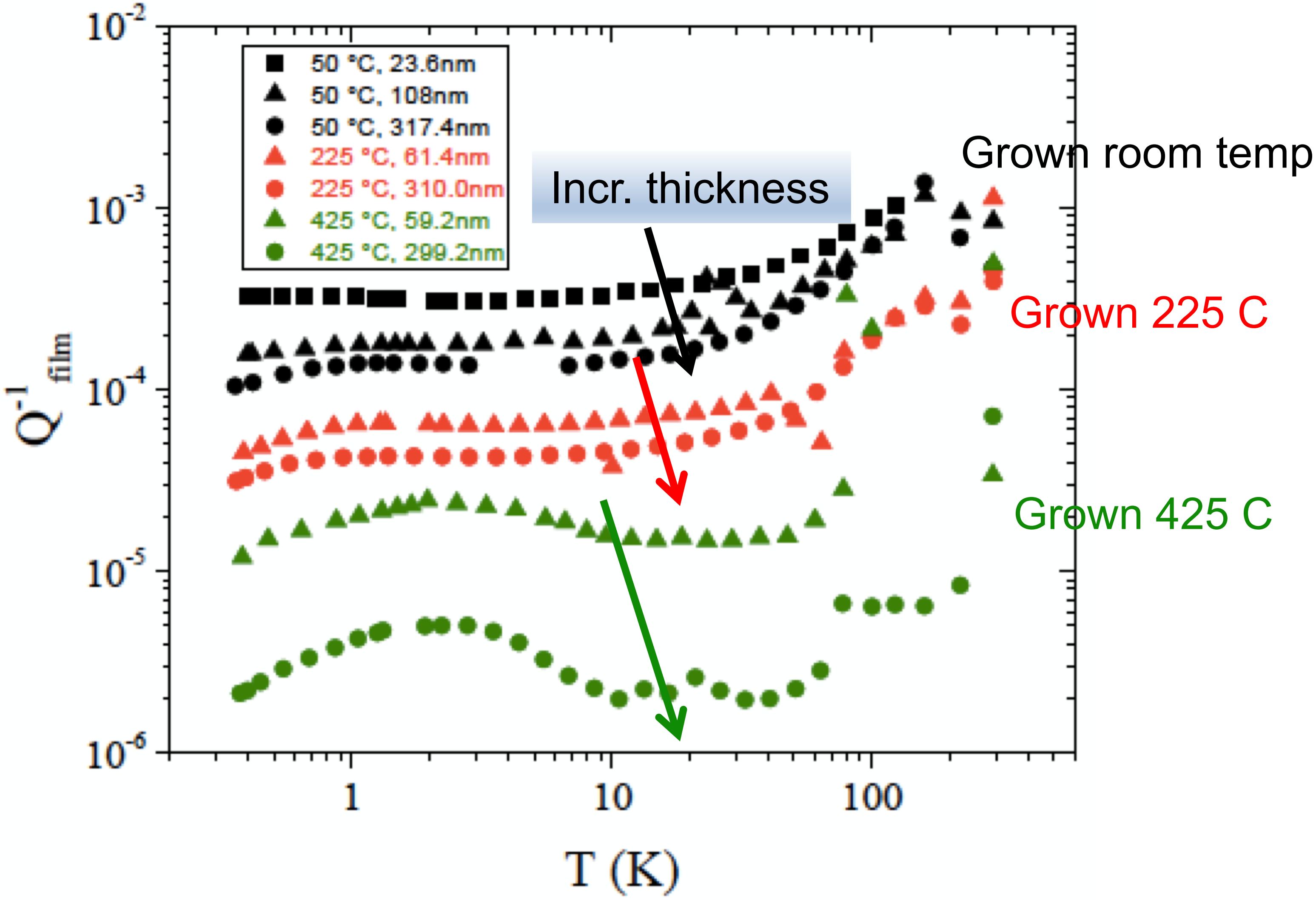
Which of these matter to TLS? Structural work **(Kiran Prasai and Hai-Ping Cheng)**

Amorphous Silicon losses: thickness also matters

Thin films are more lossy than thick films (not per volume, absolute)

Correlates with atomic density differences (thick films are denser)

Annealing reduces loss, but not much (at low T) compared to growth T effects



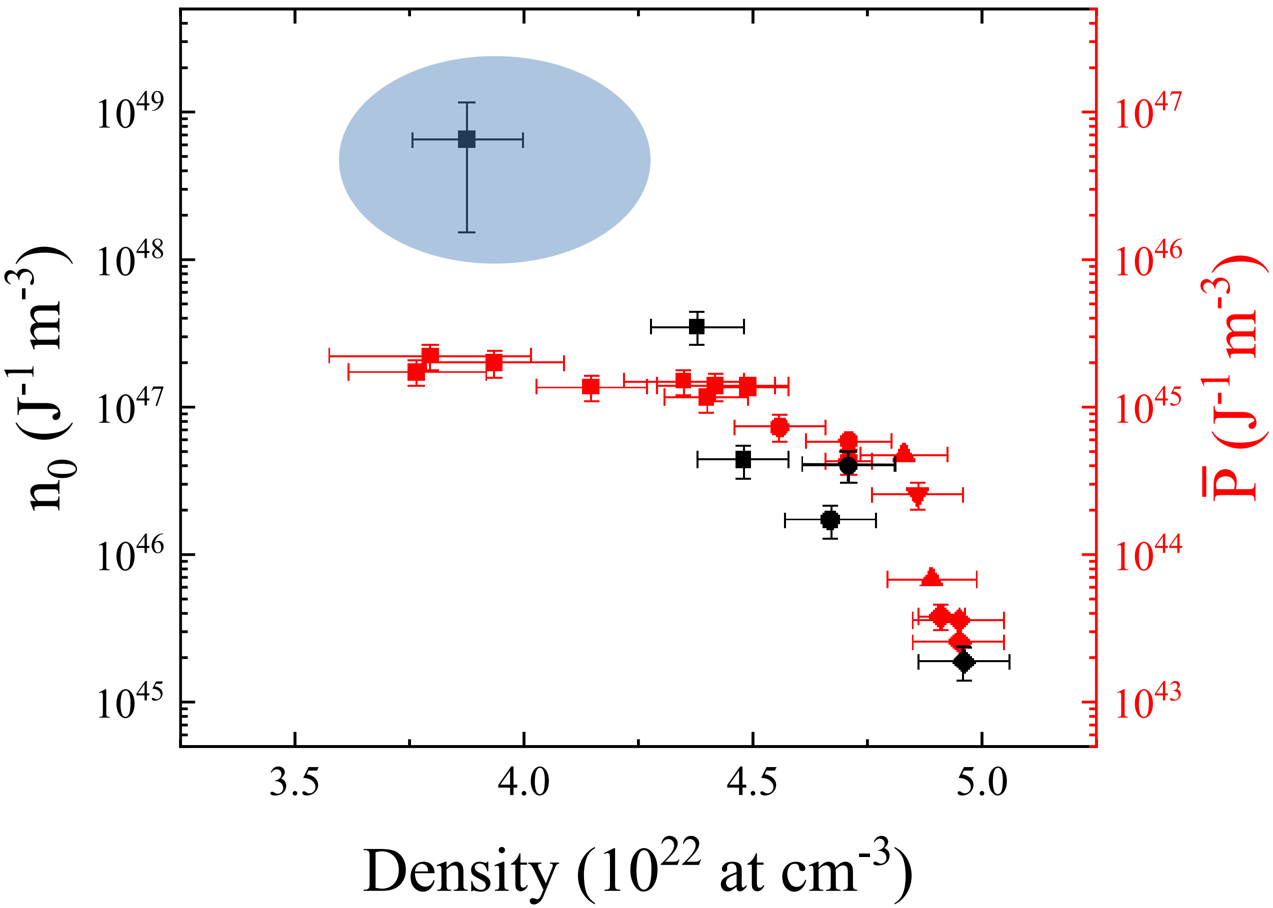
Incr. thickness

Grown room temp

Grown 225 C

Grown 425 C

More recent data on internal friction (IF) derived TLS density (specific heat still in progress)



Low density plateau in P shows that IF-derived TLS do *not* continue to increase with lower density samples

Two possible conclusions:

a) Larger nanovoids in the lower density (thinner, faster growth rate, lower growth temperature) do not create more TLS (then specific heat n_0 would also plateau)

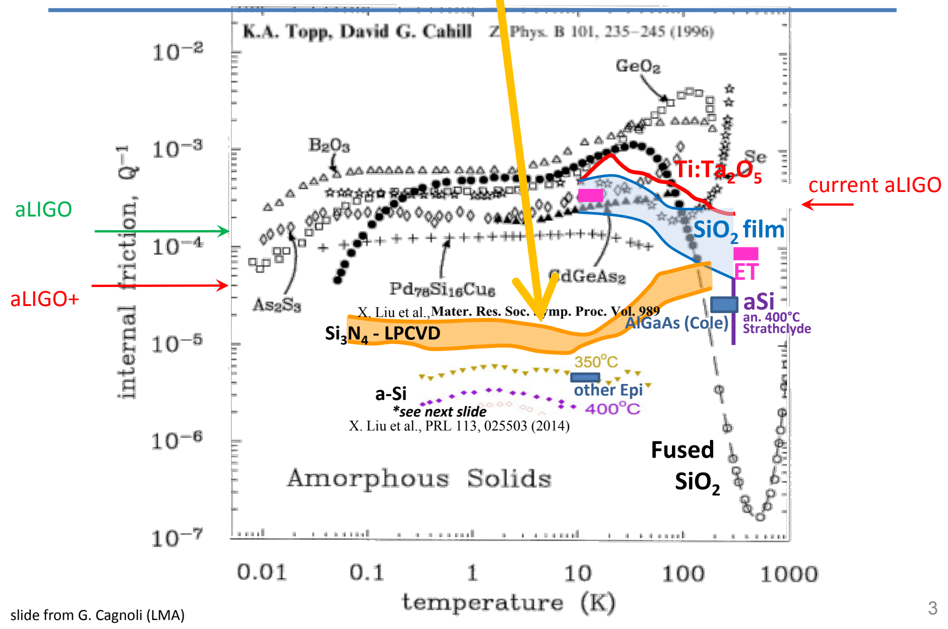
b) TLS decouple from phonons in lower density films (then specific heat n_0 would continue to increase) – one data point suggests this low γ idea

Other current materials in LIGO coatings:

Alumina (amorphous Al_2O_3) has reduced losses (at all T) when grown at elevated T_s

Amorphous Si_3N_4 – grown by LPCVD, at elevated T. Losses $<1\times10^{-4}$ at all T

Jessica, Glasgow, LVC G1800585-1

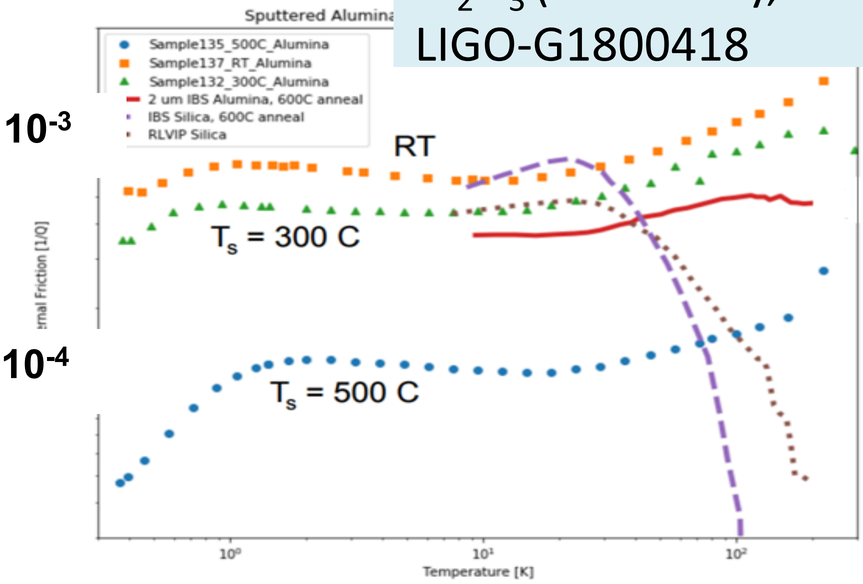


slide from G. Cagnoli (LMA)

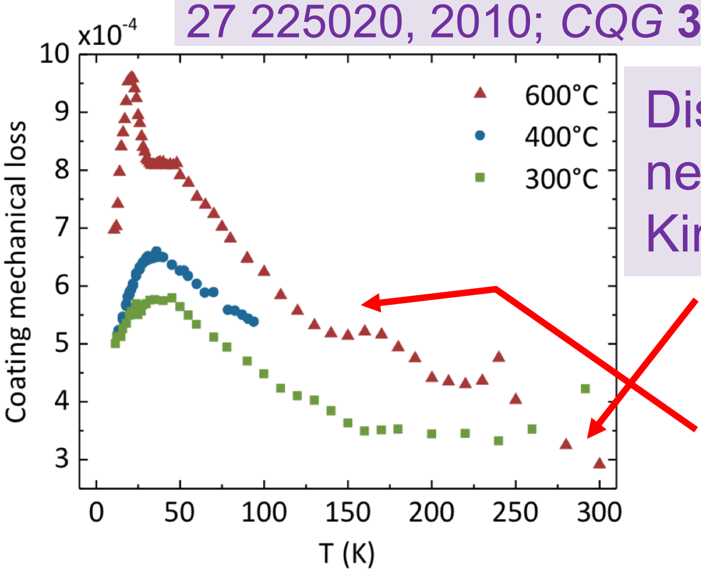
Tantala (Ta_2O_5): annealing more effective, growth T less effective – why?

- Elevated temperature growth of IBS tantala: T_s up to 400 C) reduces RT loss
- RT loss decreases with T_{anneal} until limited by crystallization

Magnetron sputtered Al_2O_3 (Abernathy, NRL) LIGO-G1800418



I W Martin et al, Class. Quant. Grav. 27 225020, 2010; *CQG* **35** 075001



Discussed in next talk Kiran Prasai

Annealing improves loss at 300K, increases it at low T

Overview and future LIGO- specific directions

Room T: Tantala is improving! Have ~1x10^{-4} loss materials via annealing & doping

- Understand better the structural causes of loss to enable further improvements (Kieran Prasai talk)
- Look for a different high index material that has bond energies like *a*-SiO_2 with really low losses. The key seems to be separation of bond energy scales
- Look for a narrow gap (high index) *fragile* glass like *a*-Si with higher band gap

Low T: Amorphous silicon is a promising *high* index material (for long λ) below 50K, with low losses (absorption still needs improvement); losses not yet known at 123K

- What about the *low* index material?
  Silica ≈ 1x10^{-4} at 123K – ok; 5x10^{-4} at 10 K – pretty high (although there is a T in the S(f,T) so maybe good enough)
  - In many cases annealing improves high temperature loss, but worsens low temperature loss. Strengthening some bonds, weakening others.
  - **Perhaps vapor deposition at elevated temperatures can access an ideal glass, low loss at low T state in SiO_2 that is inaccessible by liquid quenching or annealing! (in progress)**
- Al_2O_3 and SiN are alternative good *low* index candidates with low loss
  - May not be necessary to achieve low TLS; **it may be enough to have low coupling constant γ!** (already have seen this effect in a-SiNx, also low density *a*-Si and *a*-Si:H)