Some current developments in glassformer phenomenology: Fundamental, correlation-related, distinctions between strong and fragile glassformers, and the question of L-L phase transitions.

Austen Angell,
Chemistry Dept. Arizona State University

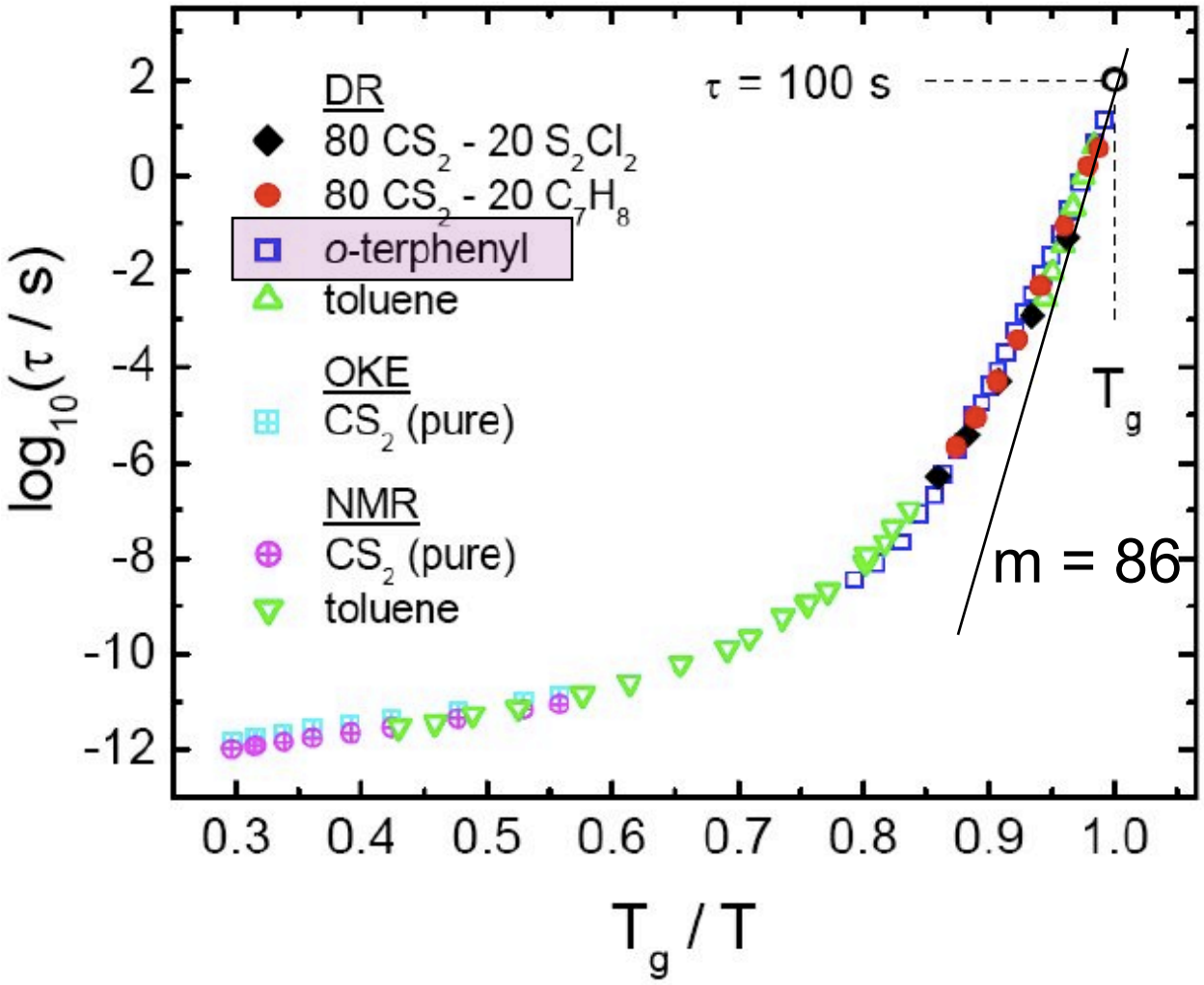
Based on work done in collaboration with Dmitry Matyushov, Vitaliy Kapko, Valeria Molinero, Srikanth Sastry, and many others.

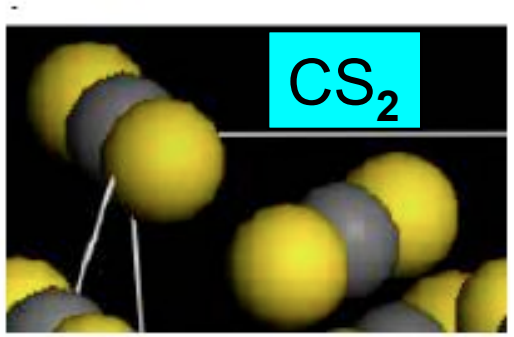
AIMING FOR THE BIG PICTURE,

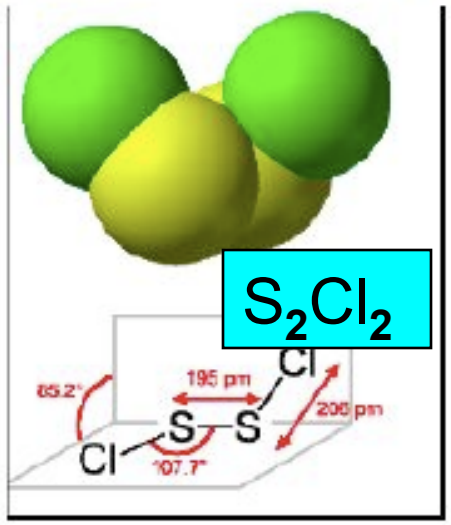
(Thermodynamic and dynamic couplings in glassforming liquids (molecular and metallic, covalent, and ionic): some new findings , some new insights)

Talk to be focused on the
metastable and stable T
ranges, i.e. T > Tg,
BUT
with predictions about what
happens below the “normal” Tg
under *equilibrium* conditions
(new resolution of the
Kauzmann paradox for fragile
liquids)

New data on fragile liquids of very simple molecular stucture







$\eta = \eta_o \exp(DT_o/(T-T_o))$

with Zuofeng Zhao and Ranko Richert, JCP 2010

Einstein observation

" If you would understand a phenomenon, but are allowed only a single type of measurement to study it, choose the heat capacity"

Here we apply this insight to…..

Outline for talk

A. BASIC CONCEPTS AND TERMS, and SYSTEMS

The glass transition for liquids- non-trivial thermodynamics

The energy landscape - and the rush to the top in some -Why the rush?

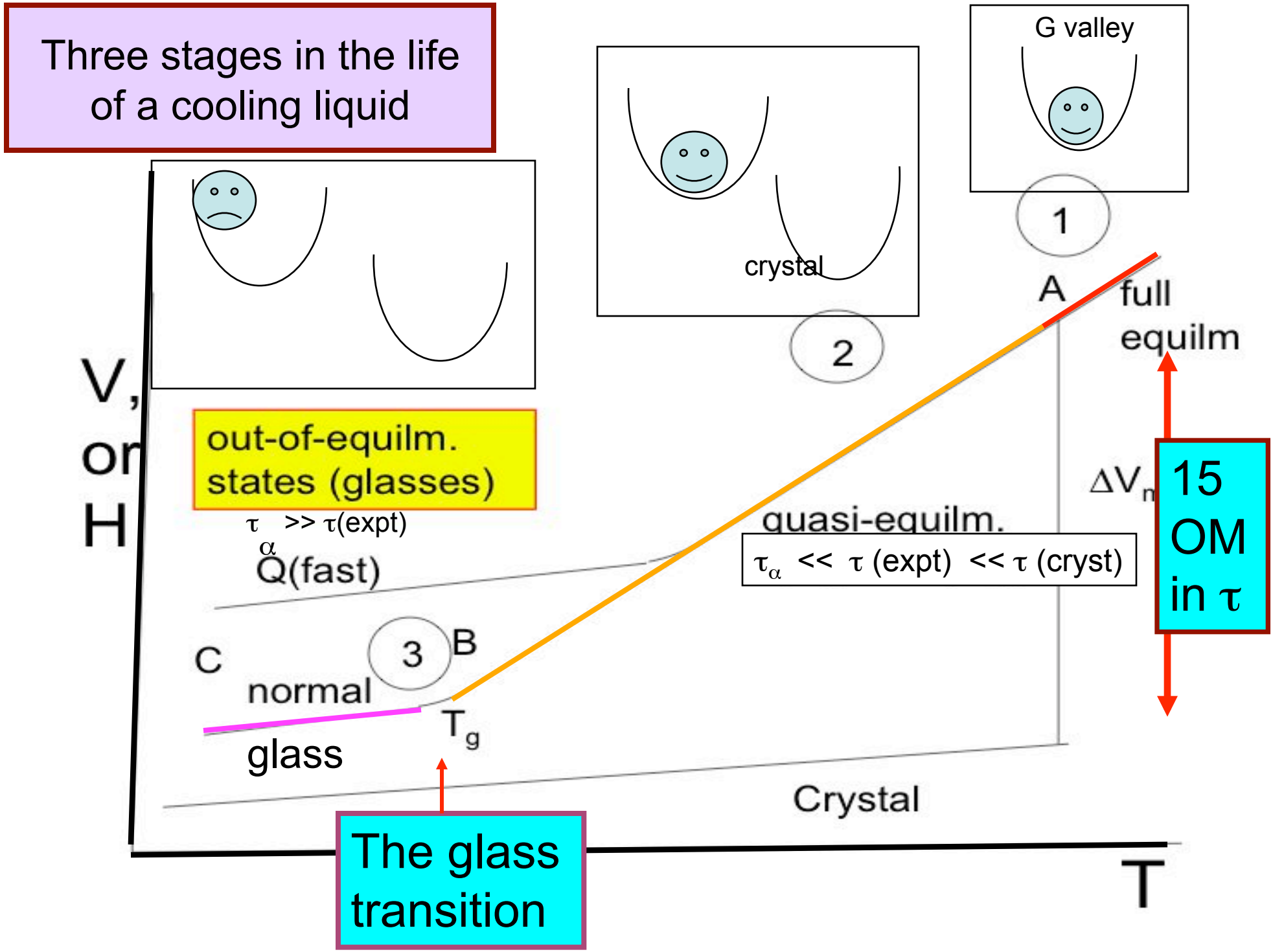
(fragile vs strong behavior)

B. HELP FROM NON-STANDARD CASES

Colloids, and crystals (rotator phases and disordering alloy superlattices) - and other λ transitions

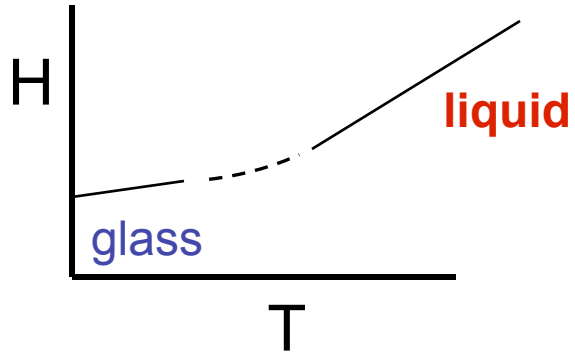
C. LIQUID-LIQUID TRANSITIONS (visible and hidden) -

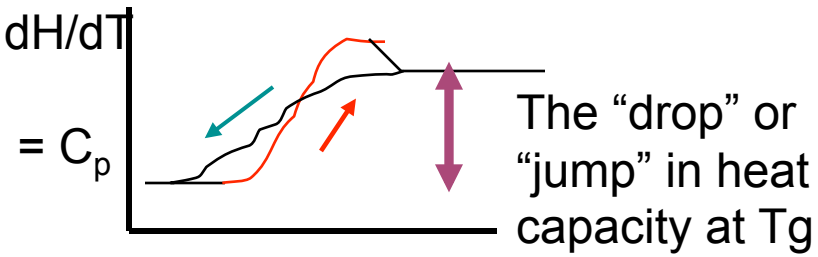
D. RELATION TO light scattering, neutron scattering, BMG phenomena, etc


Three stages in the life of a cooling liquid
G valley
crystal
1
2
3
A
B
C
full equilm
out-of-equilm. states (glasses)
τ >> τ(expt)
α
Q(fast)
quasi-equilm.
τα << τ (expt) << τ (cryst)
ΔVm
15 OM in τ
normal
glass
Tg
Crystal
The glass transition
V,
or
H
T

The GLASS transition

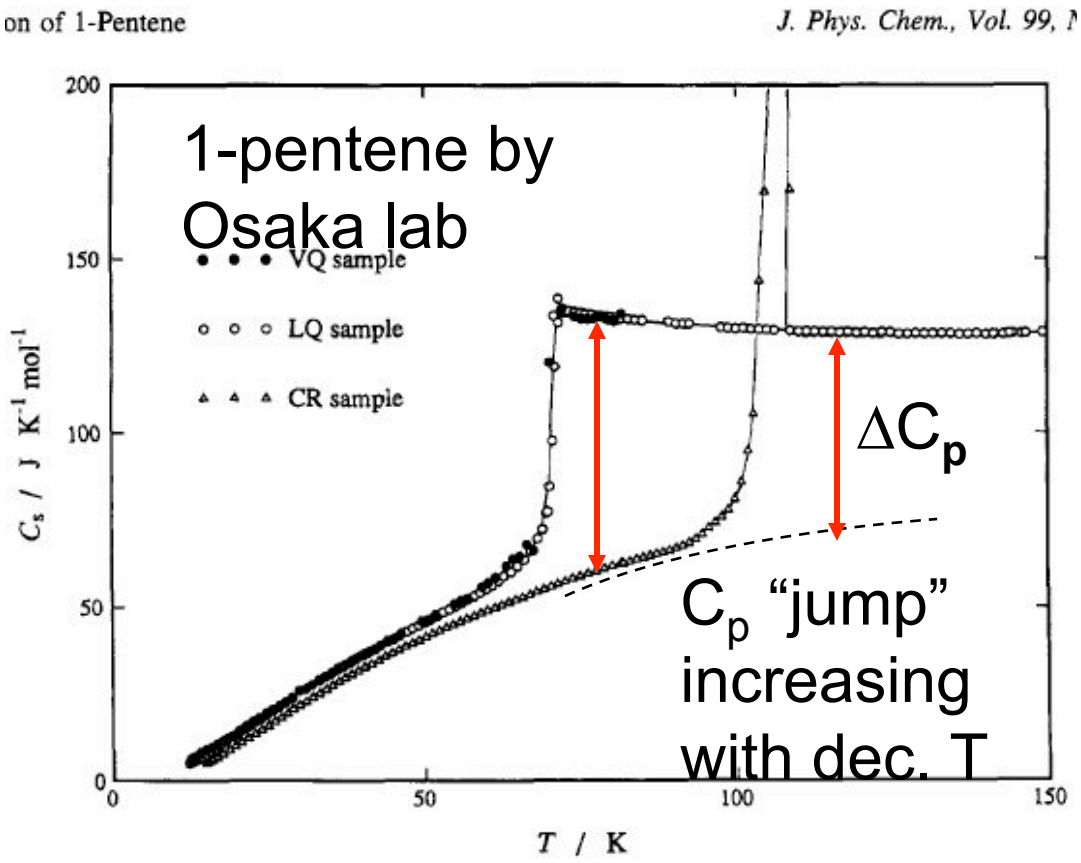
where the system gets “stuck”, “degrees of freedom” drop out, heat capacity changes





The “drop” or “jump” in heat capacity at Tg

It can be a very sudden event



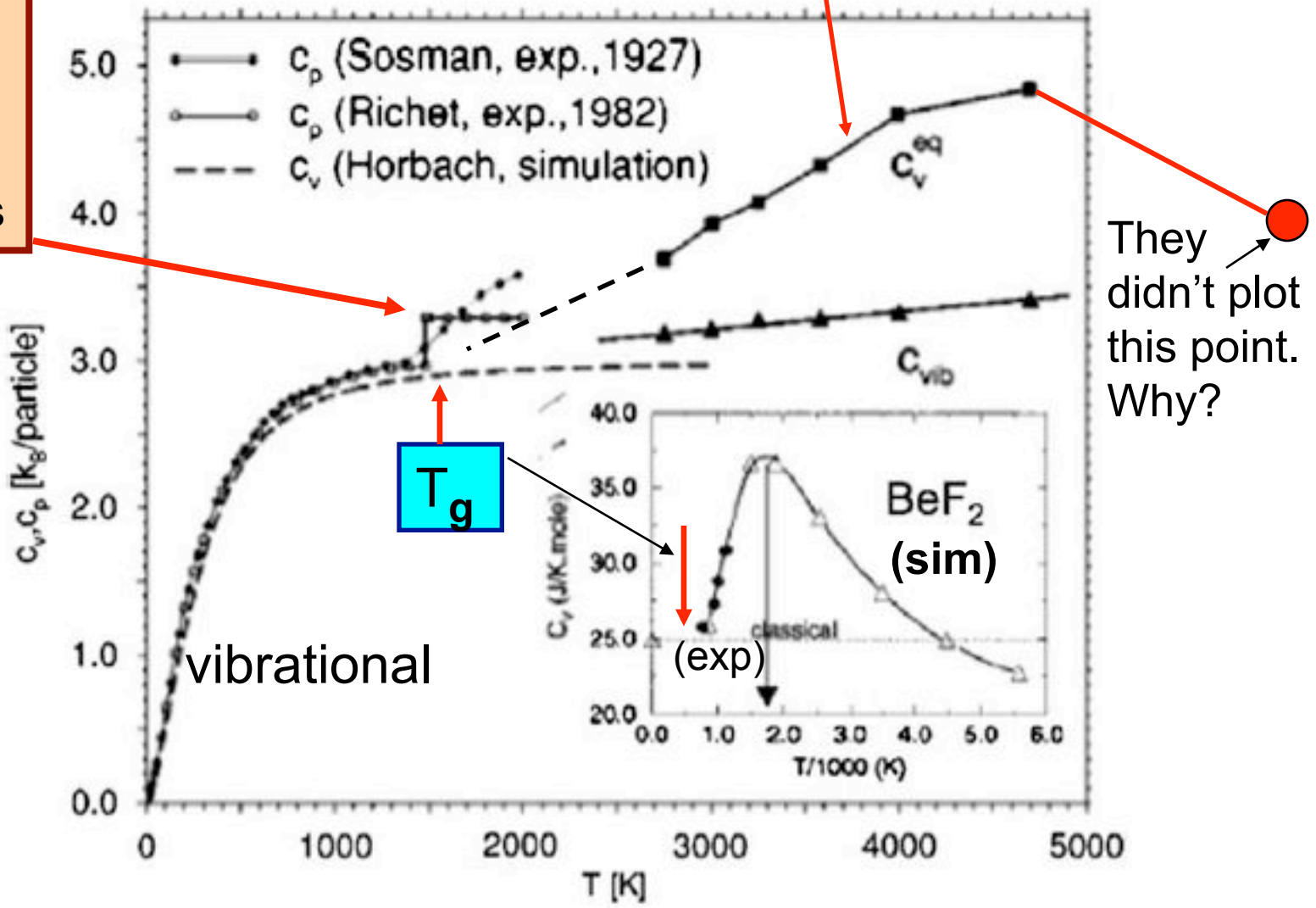
At finite T, system “floats” above some basin minimum, by vibrational excitation. At high T, floats above ToL (ToL = Top-of-Landscape). Then “simple” liquid

But also nearly almost imperceptible, e.g. dry silica

Horbach, Kob and Binder simulation "bone-dry" *(nothing in the lab so dry as a simulation with no water)*

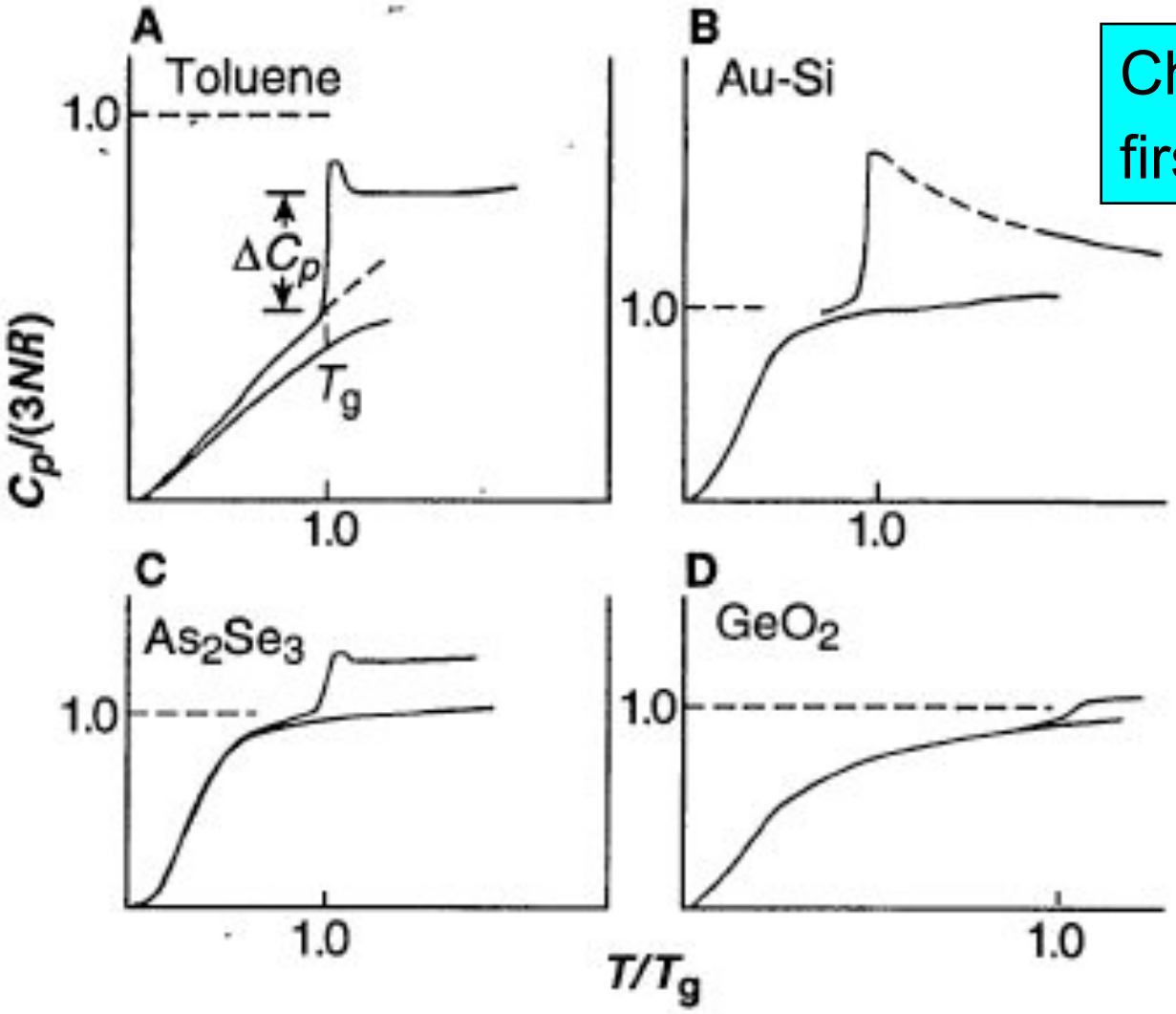
Tiny bit wet, means some broken

Si-O-Si bonds



They didn't plot this point. Why?

Heat capacities of different glassforming liquids



Chen and Turnbull first **metallic** glass

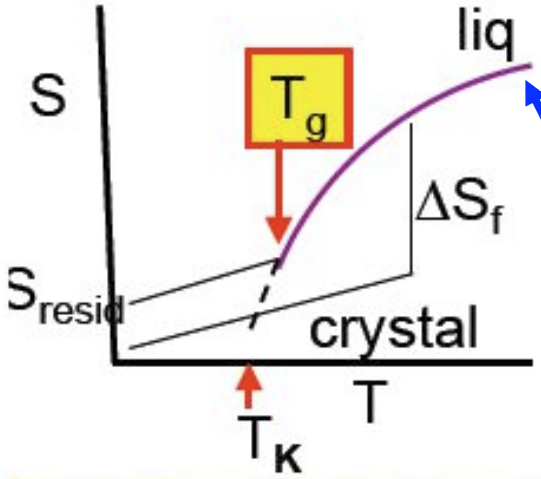
From Science, 1995, special issue on amorphous materials, (CAA)

Data can used to generate a family, by introducing the integral, S(excess)

Integral quantities..the entropy

$S = \int C_p/T dT$

$S_{ex} = S_{liq} - S_{crystal}$



S_c (ToL) ~ RlnW (per mole of **excitable units**)

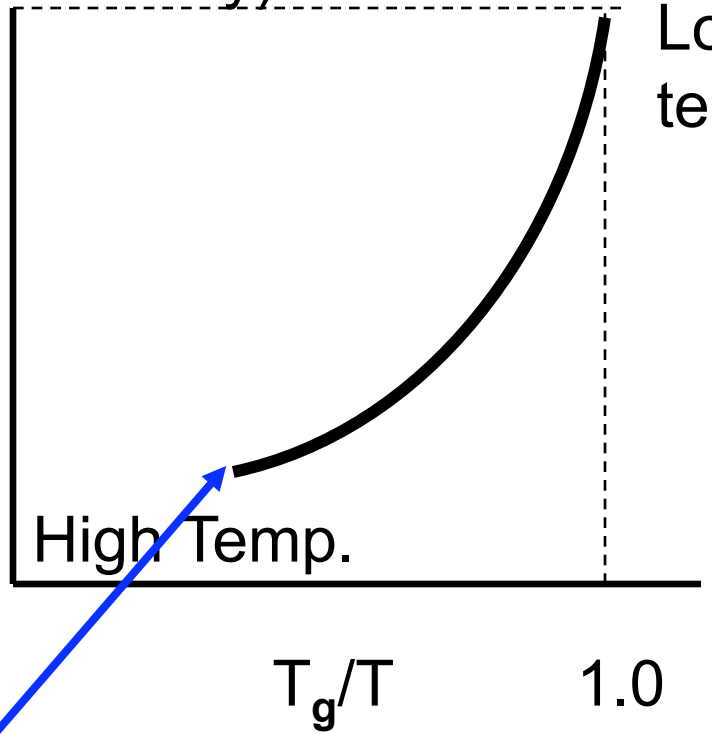
$W \sim e^N$

So S_c(ToL) **≈ R (per mole excitable units (**  **)**

Speedy, Stillinger

Plot the excess entropy in a strange scaling (wait a second for why)

$\frac{S_{ex}(Tg)}{S_{ex}(T)}$



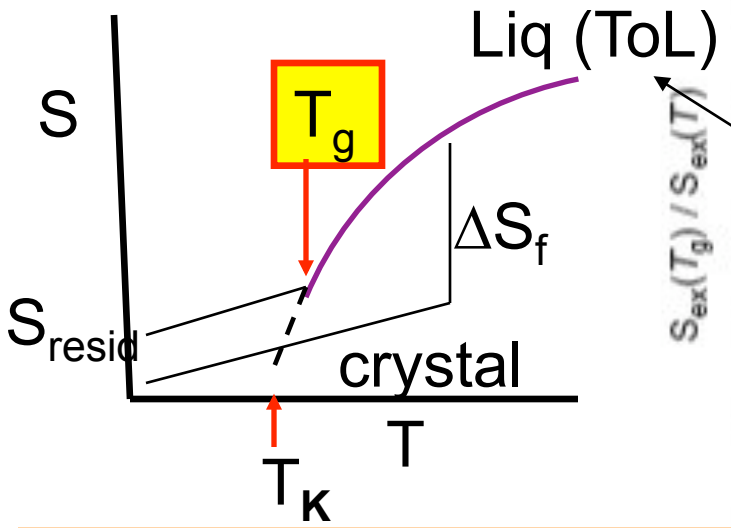
or $e^{\alpha N}$

$\alpha = 2$

T_g-scaled rate of entropy increase as $T > T_g$

$S = \int C_p/T dT$

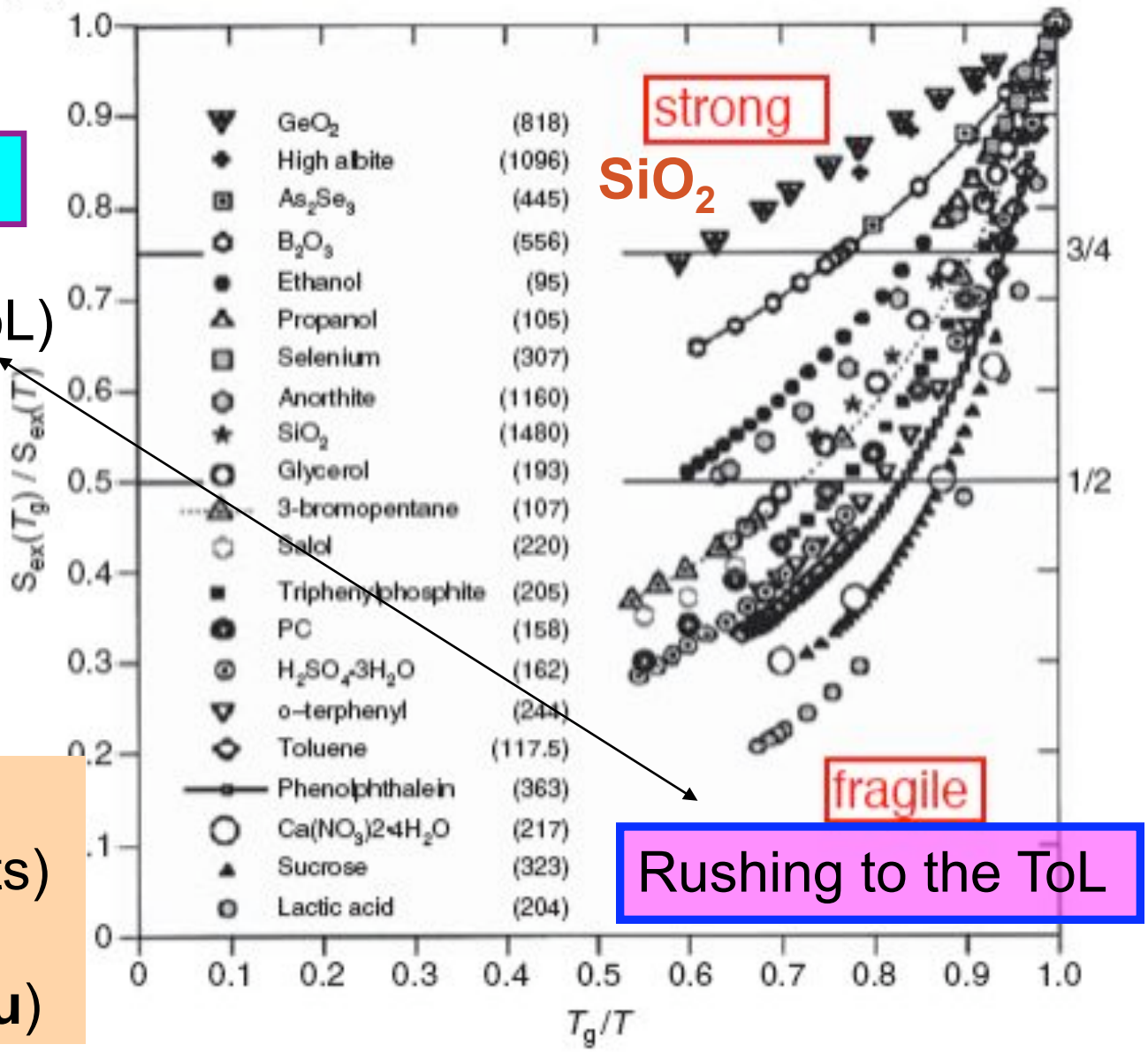
$S_{ex} = S_{liq} - S_{crystal}$



S_c (ToL) ~ RlnW (per mole of **e**xcitable **u**nits)

$W \sim e^N$

So S_c(ToL) ~ R pm(**eu**)

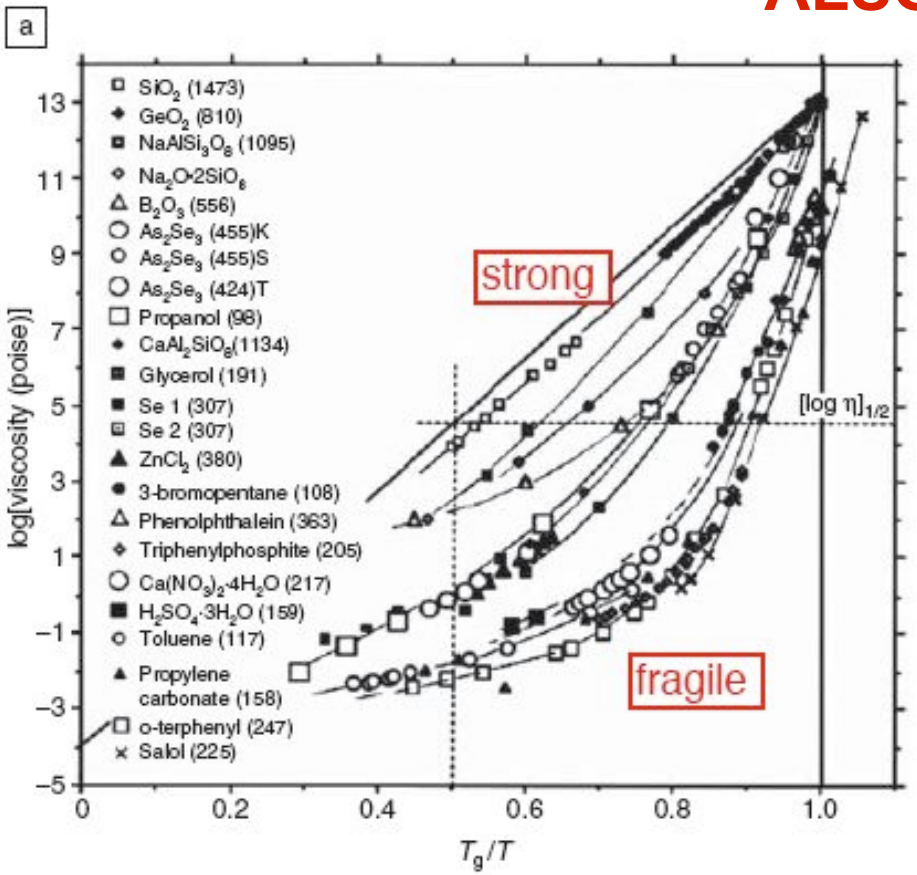


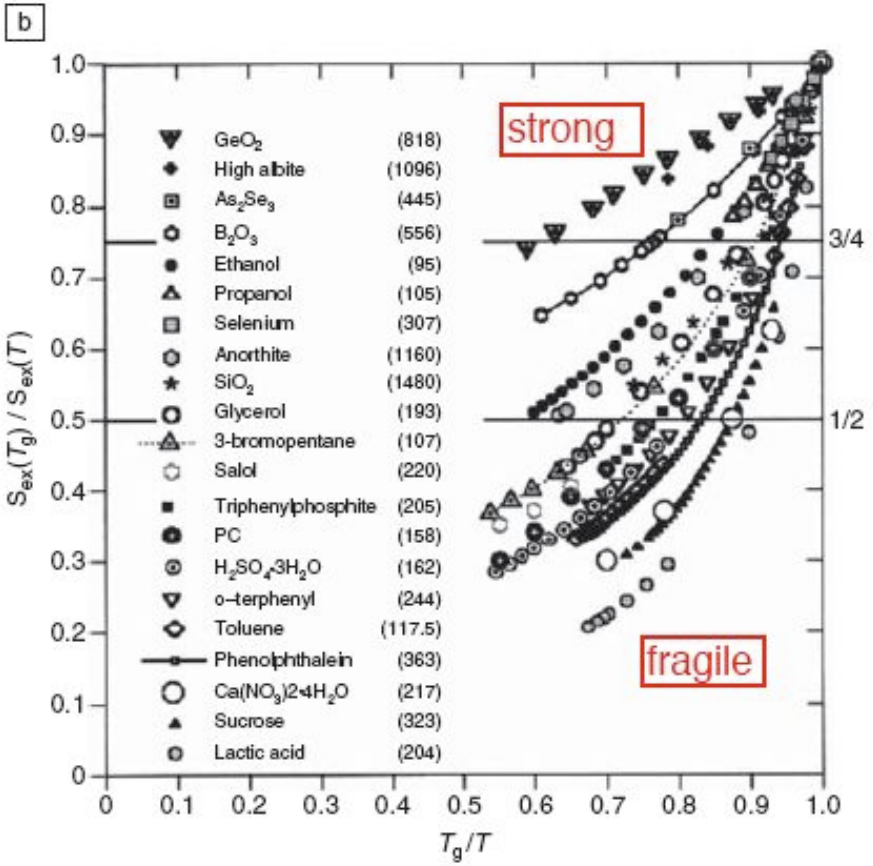
Broad sweep phenomenology

Dynamics (viscosity)

AND ALSO

Thermodynamics (entropy)

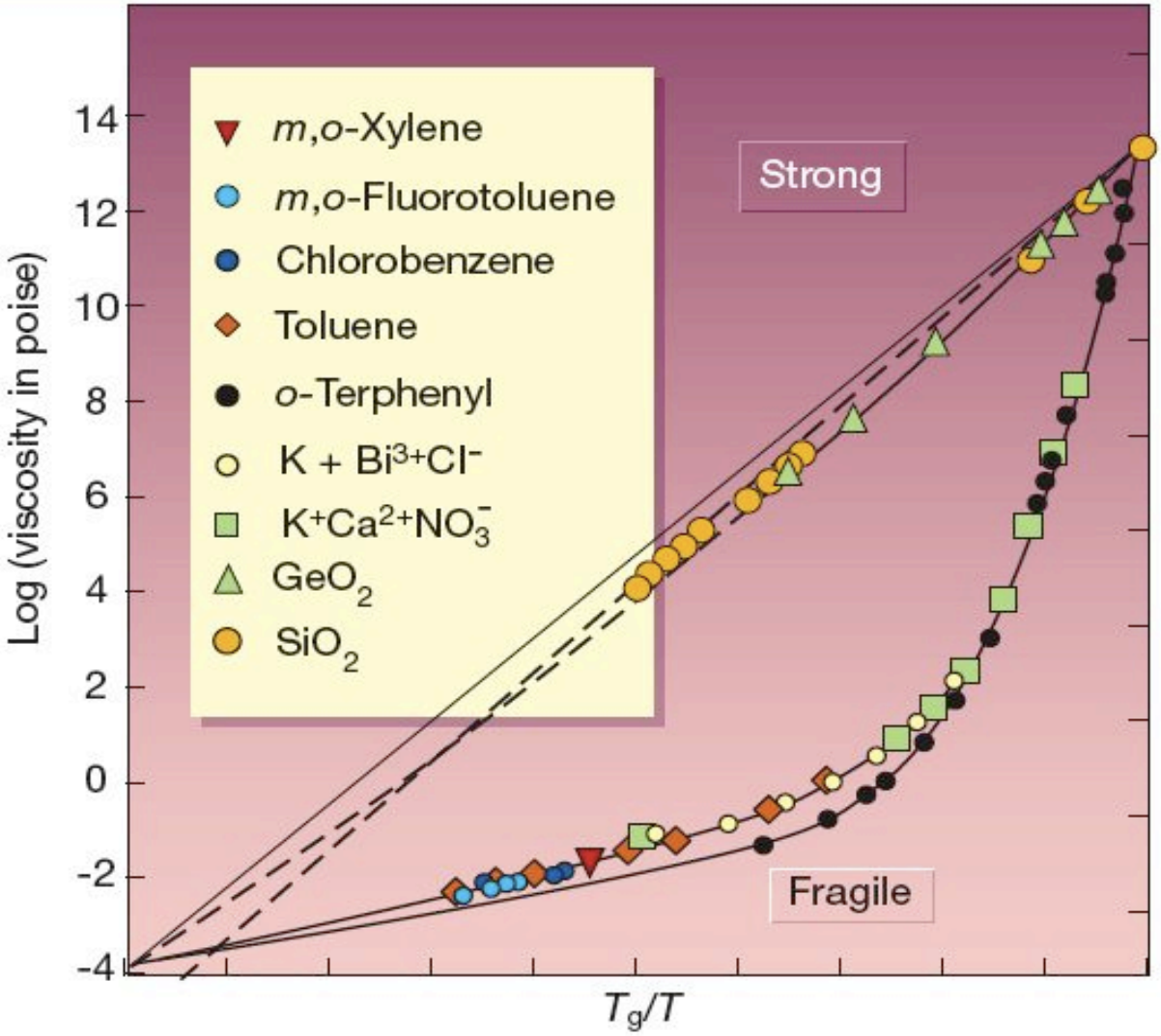




Different rates of viscosity increase

Ditto for config. entropy

Fragile liquids are striking, but *not* the archetypal glassformer **SiO_2** or its cousin GeO_2 **WOT is the difference?**



From Debenedetti and Stillinger, Nature, (2001)

Thermodynamic fragility of the same liquids

$$m_{calc} = 56\ T_g\ \Delta C_p(T_g) / \Delta H_m$$

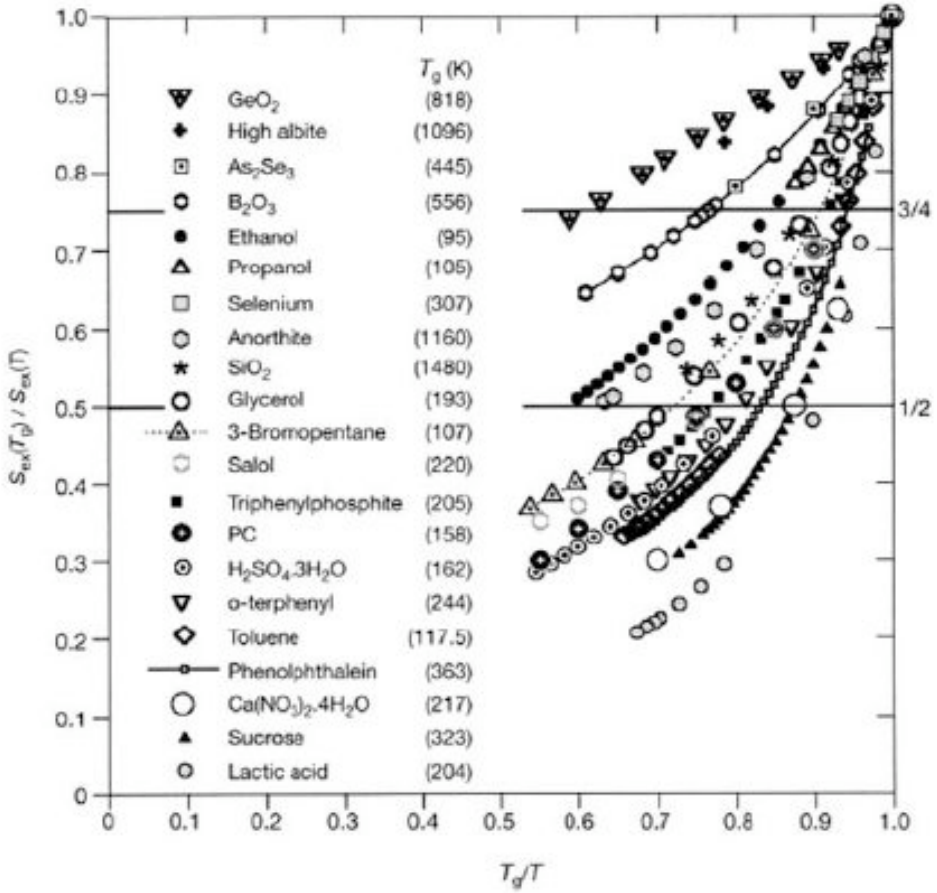


Fig. 5(b). T_g-scaled inverse temperature display of the increase of excess entropy of liquid over crystal as temperature rises above the onset glass temperature T_g.

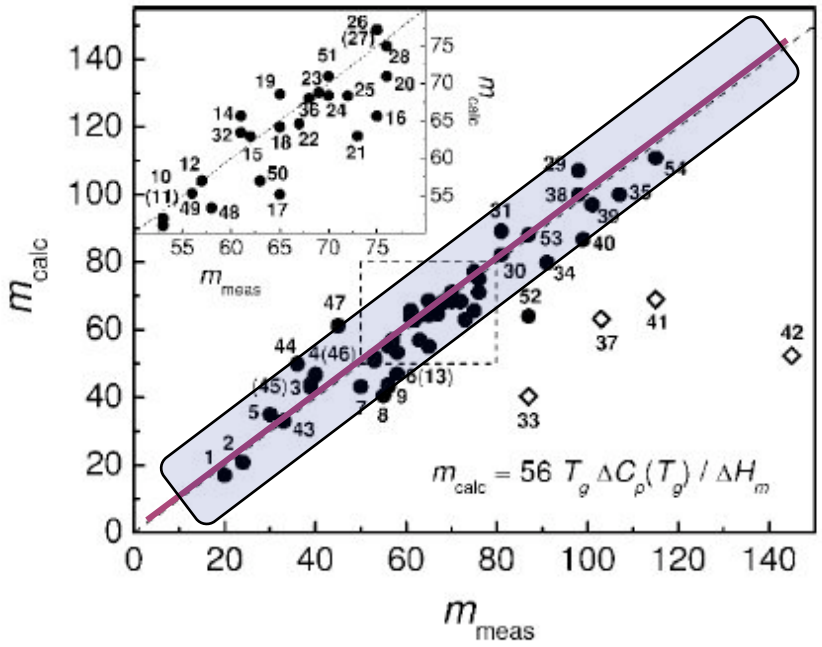


FIG. 1. Correlation of the calculated m_{calc} with the measured m_{meas} fragility index values for the 54 nonpolymeric glass-forming materials of Table I. Data in the $50 < m < 80$ range are also shown in the enlarged inset for clarity. The open diamonds (no. 33, 37, 41, and 42) are explained in the text. The dashed line represents the relation of Eq. (2), $m = 56 T_g \Delta C_p(T_g)/\Delta H_m$

So fragility is *only* correlated with large ΔC_p, if ΔS_f is normal (often confused)

Fragile liquids are liquids with structures that fall to pieces with small increases in temperature

They are “rushing” to the tops of their respective energy landscapes, driven by thirst for entropy.

What makes one liquid more fragile than the next?

It's one of the "grand challenges" of the glass problem.

But no time for it here

Instead we ask……Are strong liquids (glassformers) and fragile liquids just extremes, or are they opposites?

We are interested in the nature of fluctuations and their correlations, both static and dynamic,

e.g. (1) density (and dielectric constant) fluctuations responsible for **compressibility** and scattering of liqht

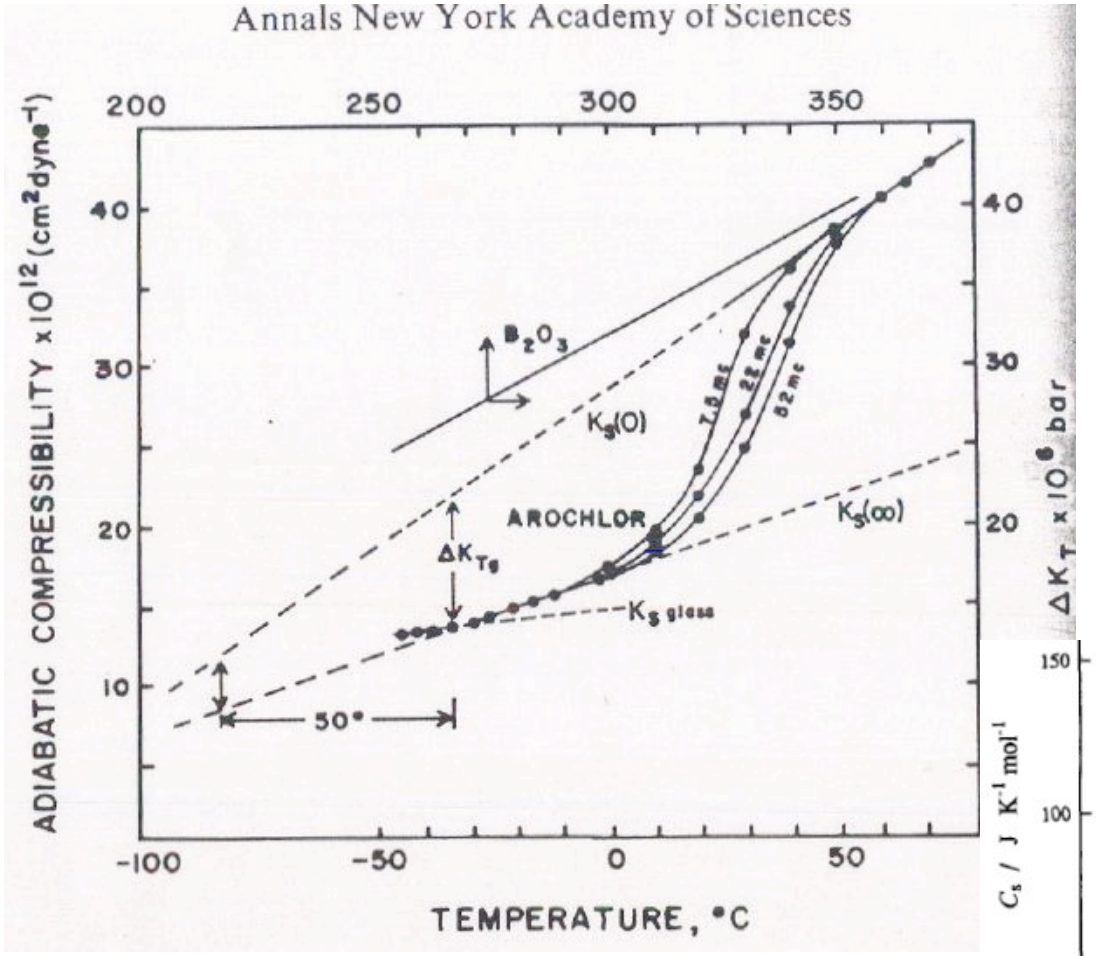
and (2) enthalpy fluctuations that determine **heat capacity**

Landau and Lifschitz “Statistical Physics”

$$\kappa_T = \langle \Delta V^2 \rangle / VkT$$

$\langle \Delta V \rangle$ goes as inverse of density fluctuations

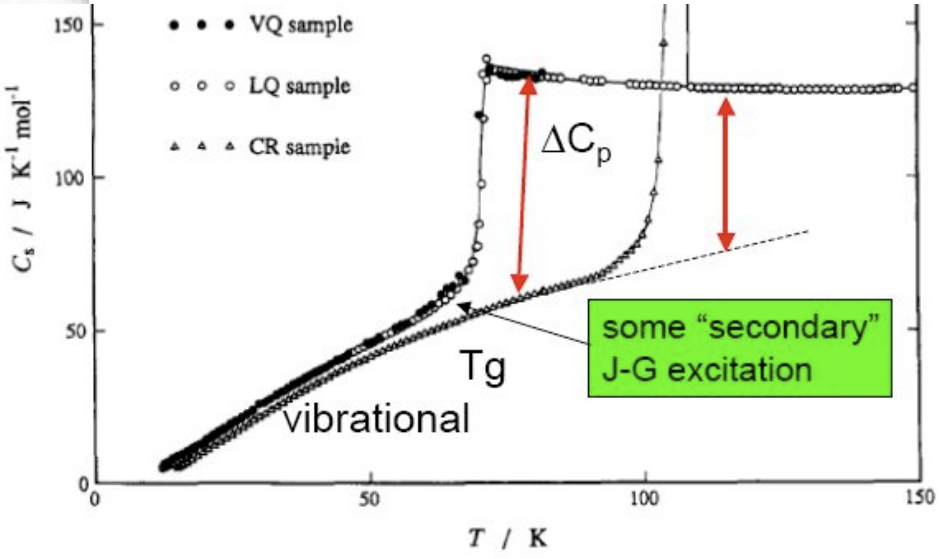
The excess compressibility of supercooled liquids is little studied, but seems to be decreasing with decreasing T



(from Litowitz et al)

Contrast with excess heat capacity

~ $\langle \Delta S^2 \rangle$



Compressibility of OTP (fragile) vs T

Naoki and Koeda,
J. Phys. Chem.
93, 948, 1989

$\Delta\kappa_T$ is decreasing with decreasing T! Compare this with expansivity and heat capacity behavior

So, entropy fluctuations, but not density fluctuations are important in fragile liquids

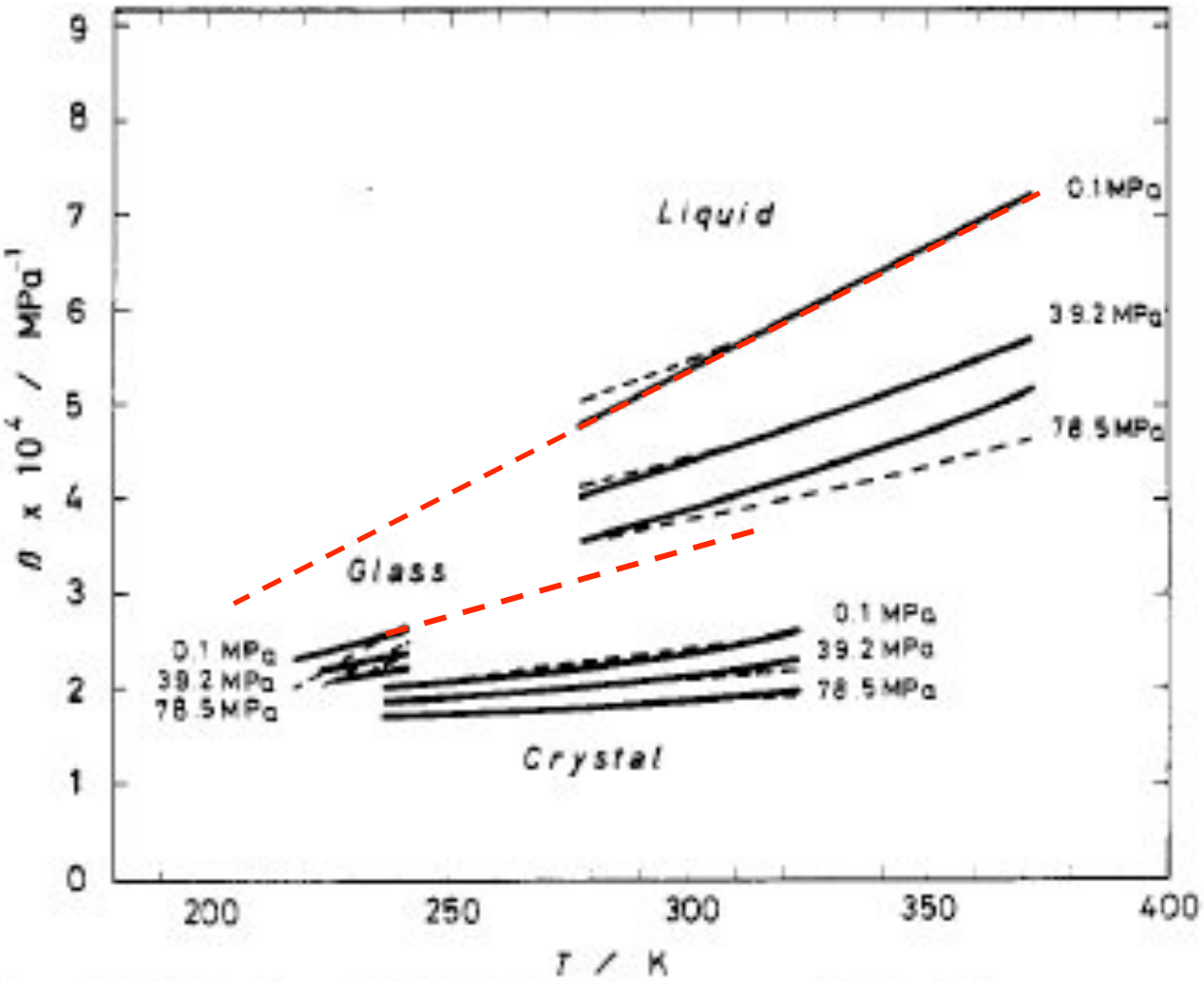
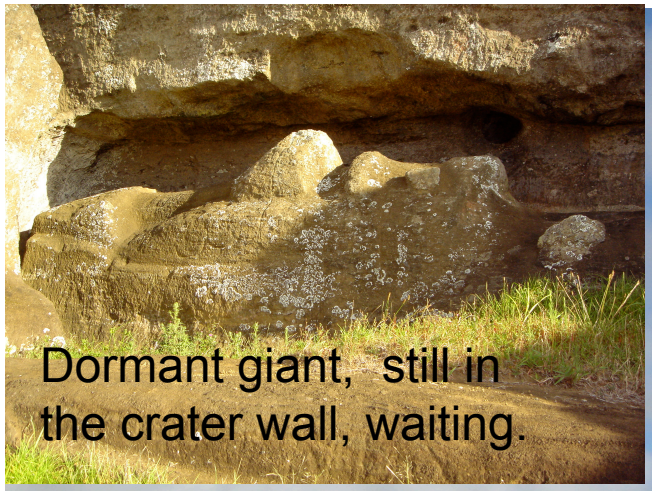


Figure 5. Variations of the isothermal compressibility with temperature. The solid lines are representative isobars calculated from eq 1 and the broken lines are the ones calculated from the thermal–pressure coefficient and the thermal expansivity.

Silent watch over Easter Island



Dormant giant, still in the crater wall, waiting.

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Colloids, and crystals (rotator phases and disordering alloy superlattices) - and other λ transitions

C. LIQUID-LIQUID TRANSITIONS (visible and hidden) -

D. RELATION TO light scattering, neutron scattering, BMG phenomena, etc

Correlations: Static or dynamic

Some say, static correlations are not important in glassformers. Rather, one must look at dynamic correlations, suggested by dynamic heterogeneity (associated with the nearly universal non-exponential relaxation functions)

Others say, including us, but particularly
Tanaka et al, in Nature Materials, Feb. 2010,
that static correlations are of overarching importance and the rest is "exotic physics". But one must look for the right correlations..**density** correlations are **not** the thing to look for. ($\Delta\kappa_T$ vs ΔC_p, and Adam-Gibbs vs free volume)

Tanaka et al use COLLOIDAL glassformers, in their studies

Background tips on colloidal glassformers

• colloidal particles are suspended in a small-molecule fluid

•Colloidal particles maybe hard-sphere like and not absorb solvent
or,
They may be spongy and change size with temperature by absorbing solvent

•Hard sphere colloids, act like hard sphere liquids, *with packing fraction **PF** being the important variable* (see next slide, where we show this behavior is extreme fragile)

•Soft sphere colloids can distort under pressure of packing and their behavior turns out to be like “strong” liquids

•Hard sphere colloids jam at **PF** 0.58, crystallize in a favored PF range, and are stabilized against crystallization by polydispersivity

Hard polymer colloids are fragile liquids

MATERIALS SCIENCE

Soft is strong

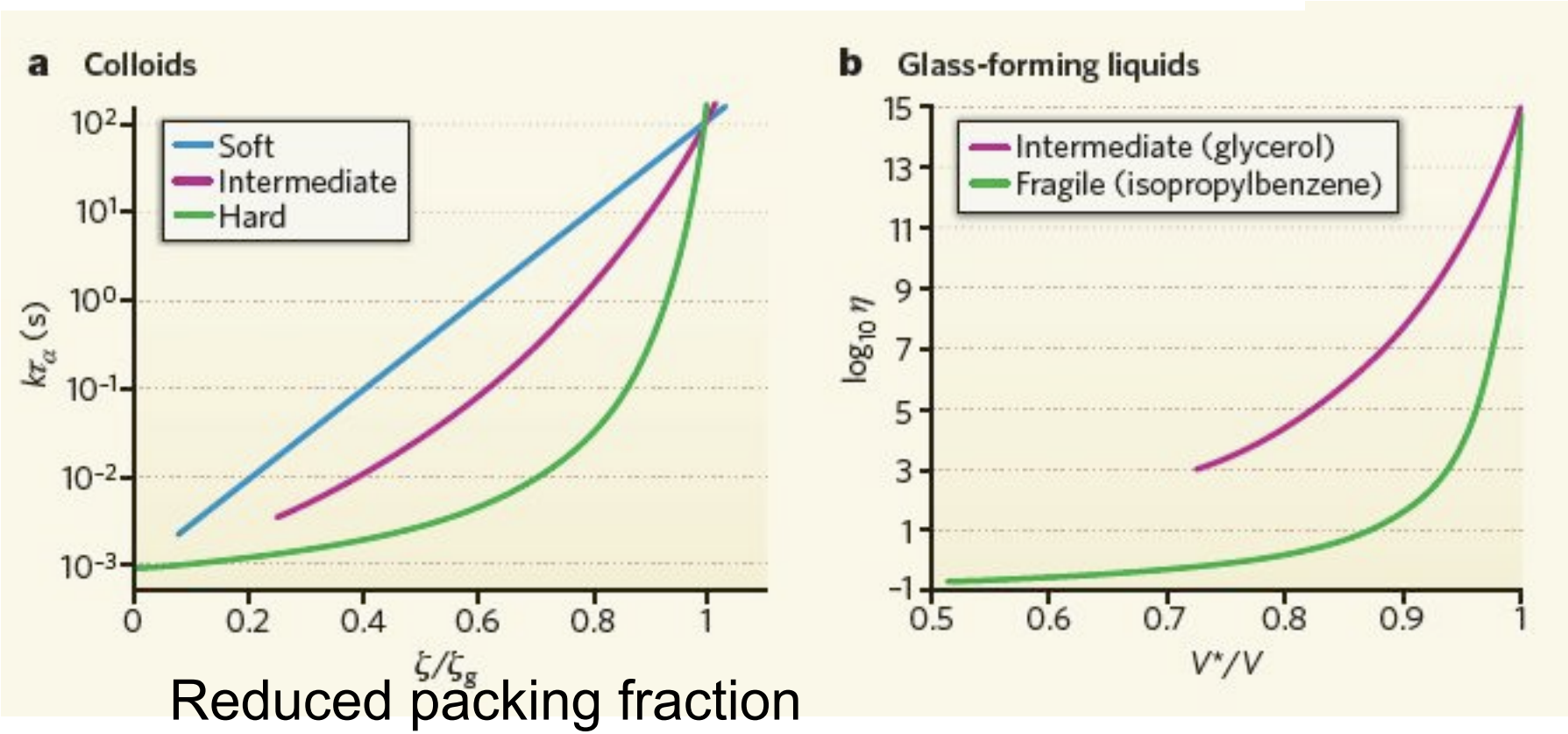
NATURE|Vol 462|5 November 2009

NEWS & VIEWS

C. Austen Angell and Kazuhide Ueno

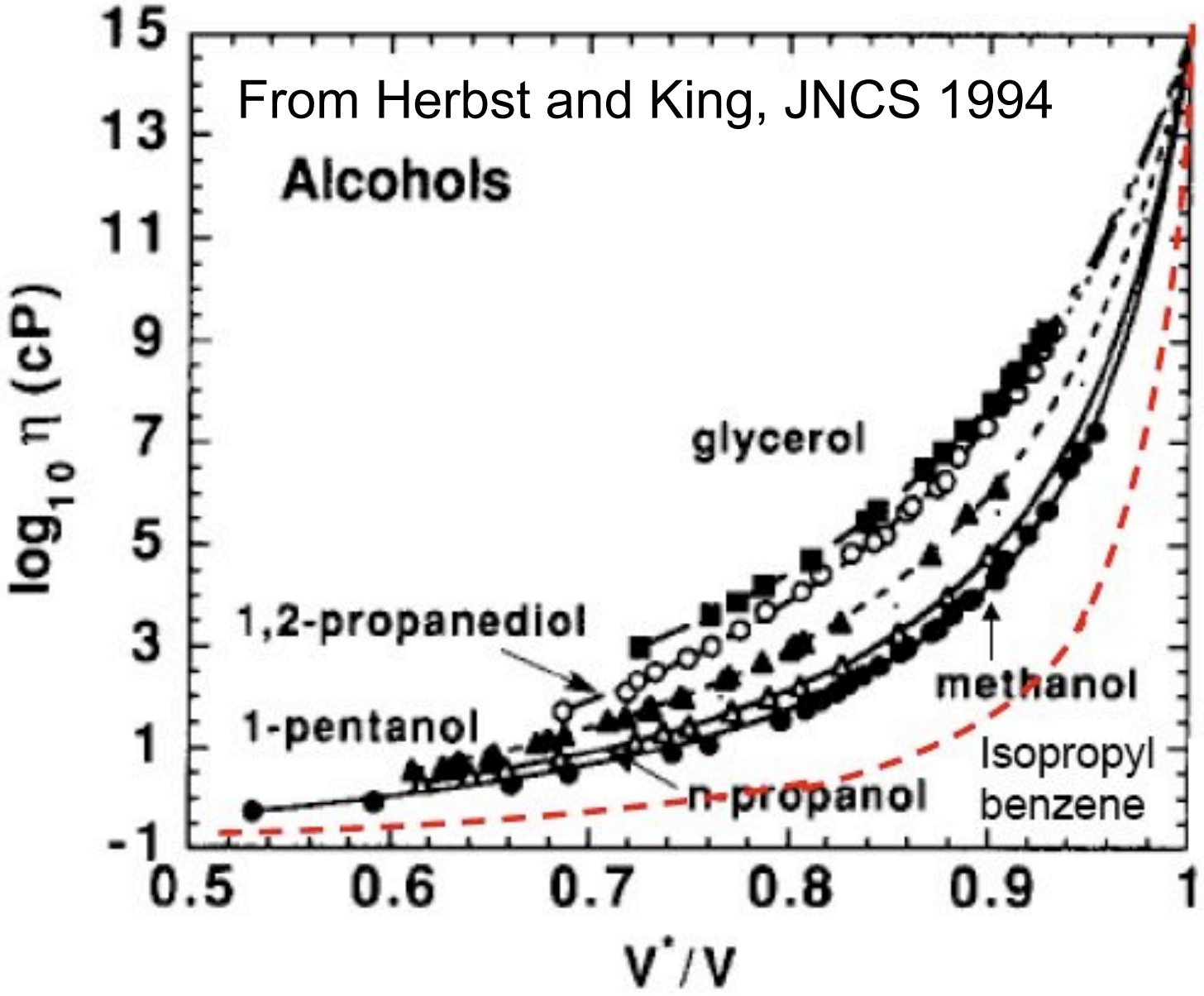
On a Nature letter by Mattson et al (Weitz)

The mechanisms that govern the rate at which glasses soften on heating have long been a mystery. The finding that colloids can mimic the full range of glass-softening behaviours offers a fresh take on the problem.



Reduced packing fraction

Molecular liquids: viscosity vs scaled volume



Enter the static correlation length

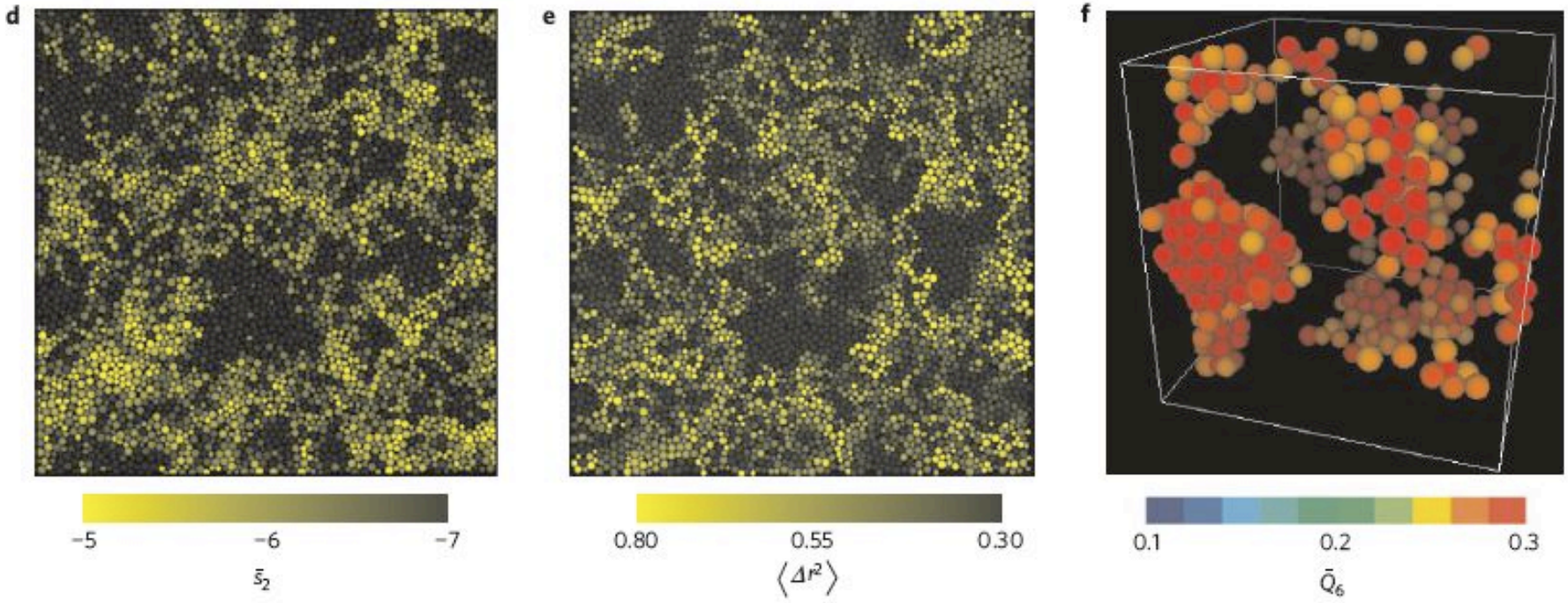
nature materials

ARTICLES

PUBLISHED ONLINE: 21 FEBRUARY 2010 | DOI:10.1038/NMAT2634

Critical-like behaviour of glass-forming liquids

Hajime Tanaka*, Takeshi Kawasaki, Hiroshi Shintani and Keiji Watanabe

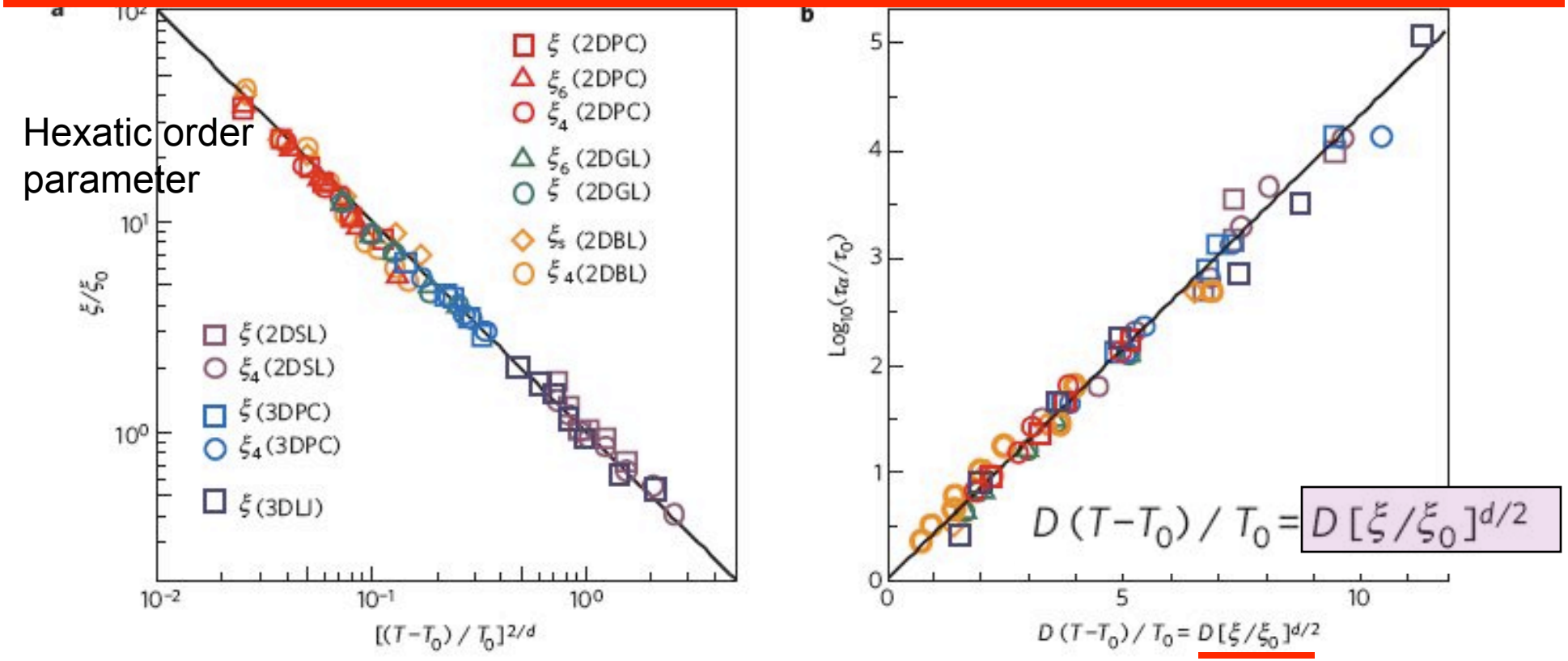


Hexatic order parameter

ure 1 | Structural order in glass-forming liquids. a, Spatial distribution of $\bar{\Psi}_6$ for 2DPC with $\Delta_{2DPC} = 9\%$ at $\phi = 0.740$. See Supplementary Video S1 for

Growing order and slow dynamics

Note relaxation times go with the correlation **length** (for MRCO)



gure 4 | The growth of medium-range crystalline order and its relation to the slow dynamics. a, Relation between ξ/ξ_0 and t^* for all of the systems:

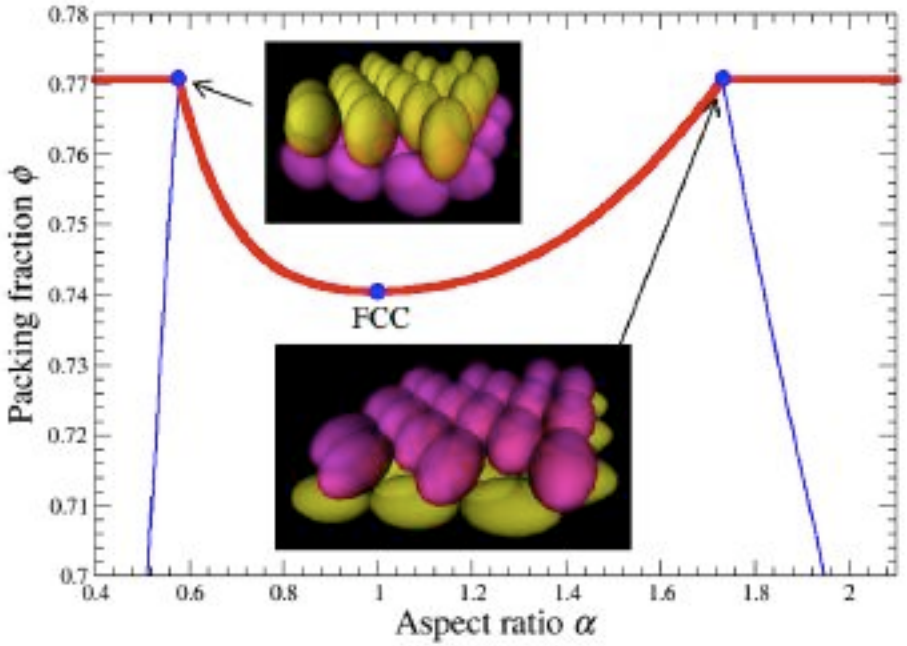
future. These results suggest that the behaviour of glass-forming liquids can be explained by a critical-phenomena-like scenario in which the correlation length of MRCO diverges towards the hypothetical ideal glass-transition temperature, T_0. We note that τ_ξ is the lifetime of dynamic heterogeneity, which may be the slowest timescale of the system.

(aside)

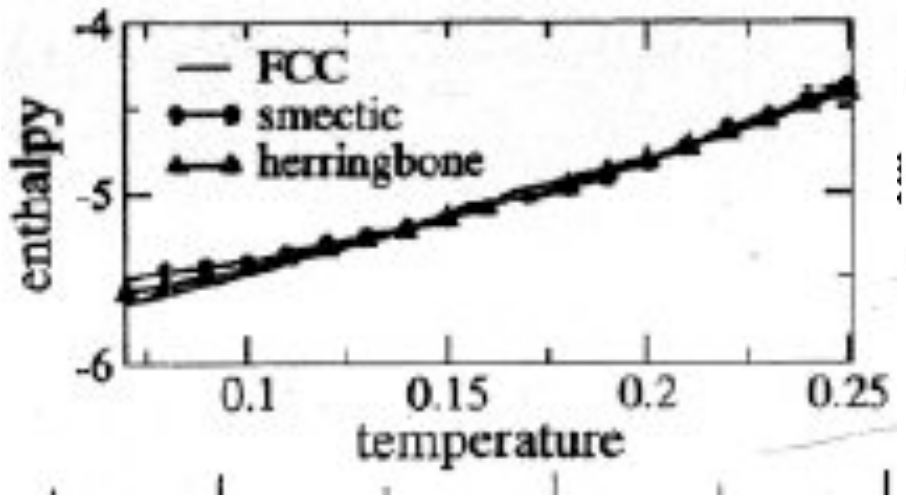
But it need not be crystalline order

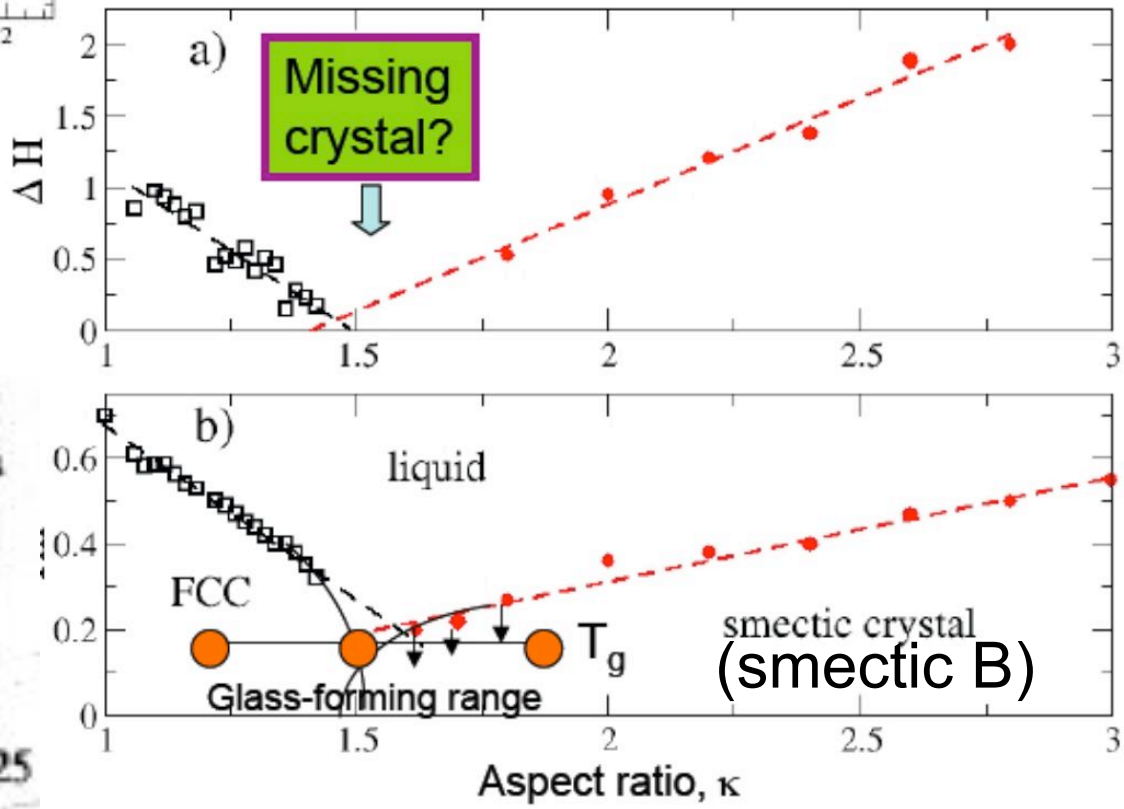
(”ideal glassformers”)

Study of the Gay-Berne model (Vitaliy Kapko)



Hard ellipsoids,
(Donev et al, PRL, 2004





(Continuing on the static correlations question)

Need some extra help from "outside"..this time on the "strong" side of the liquid scenario

We now look at "glass" transitions in non-liquid systems.

Firstly, rotator phases generally called "plastic crystals"

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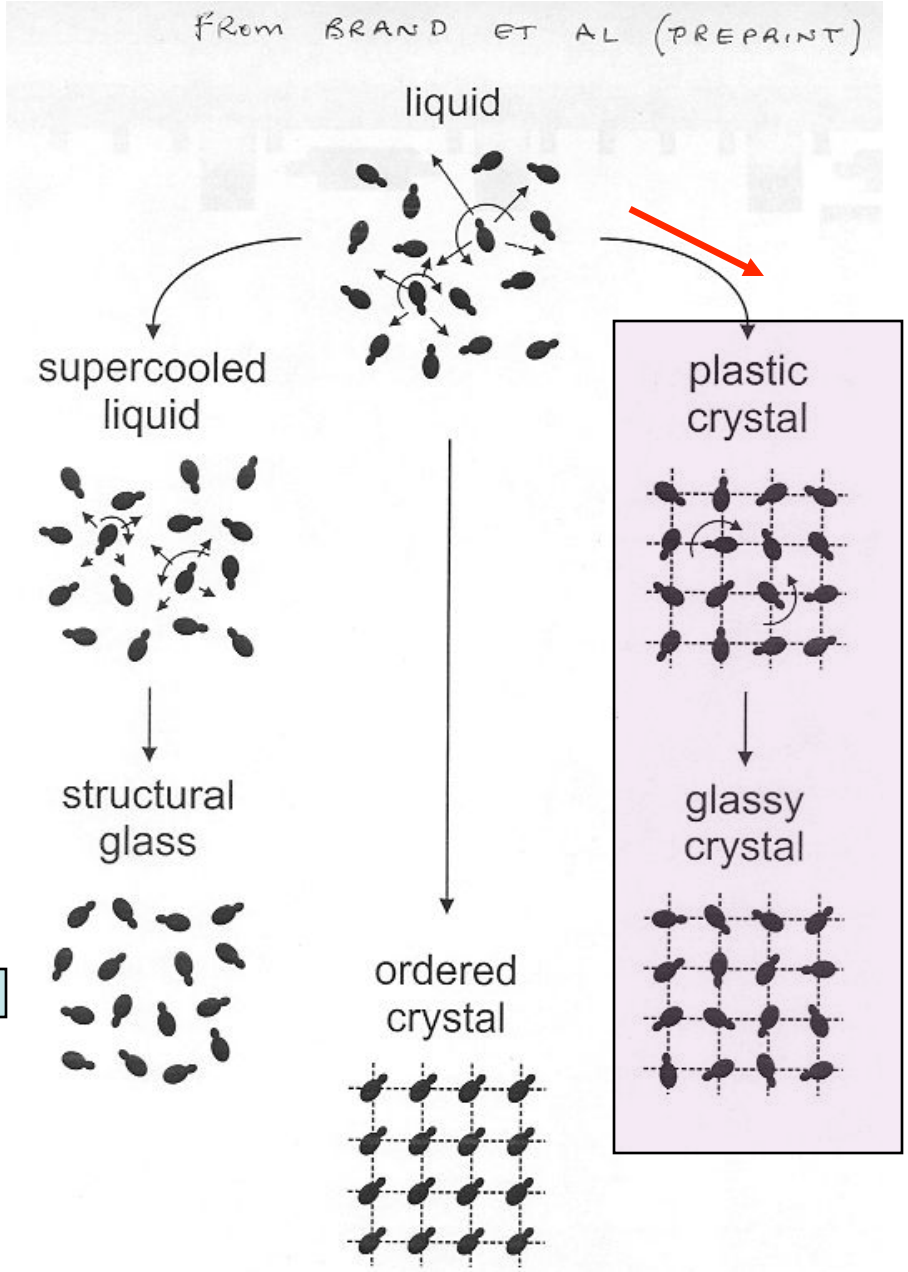
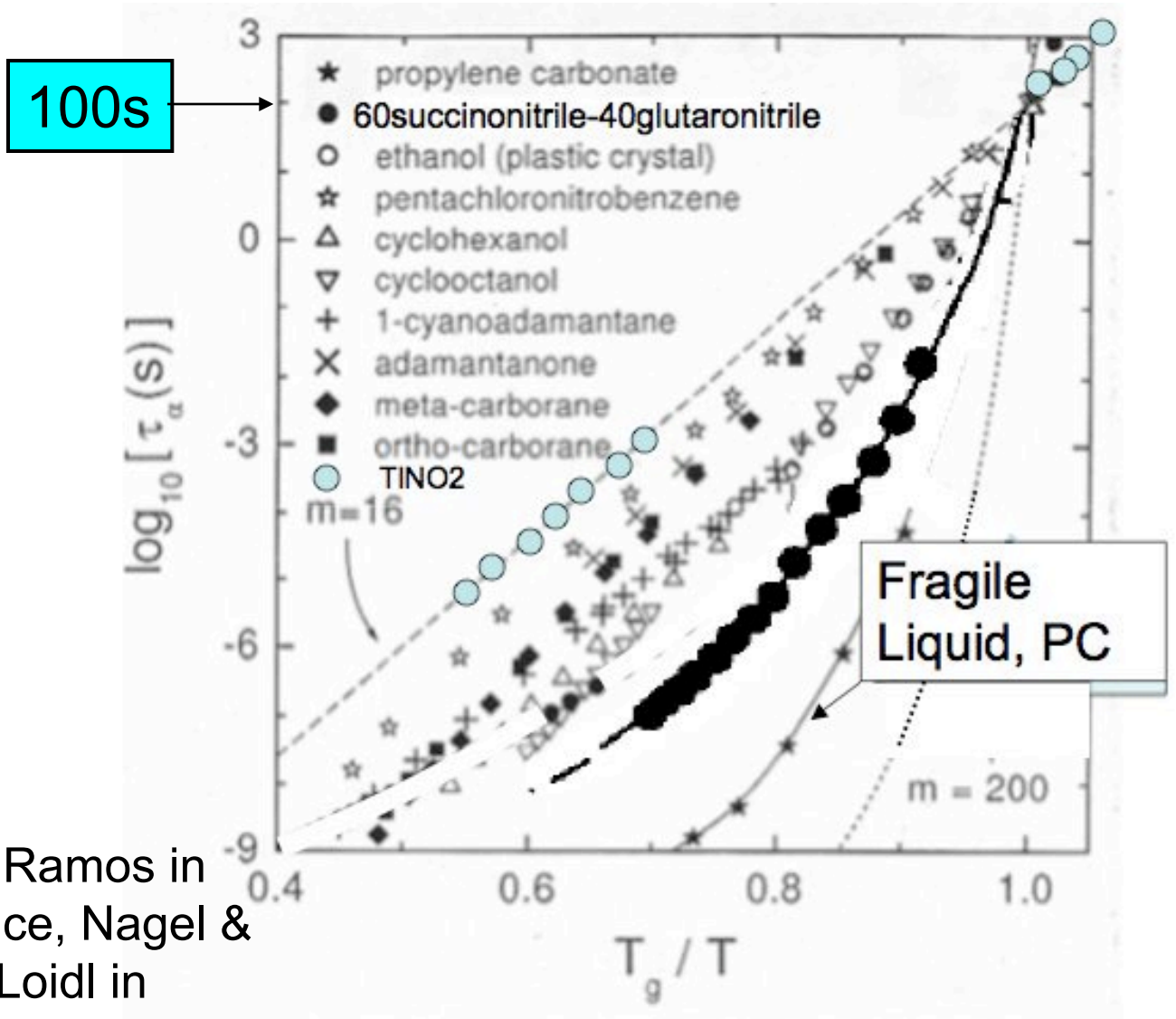
from Brand Lunkenheimer and Loidl



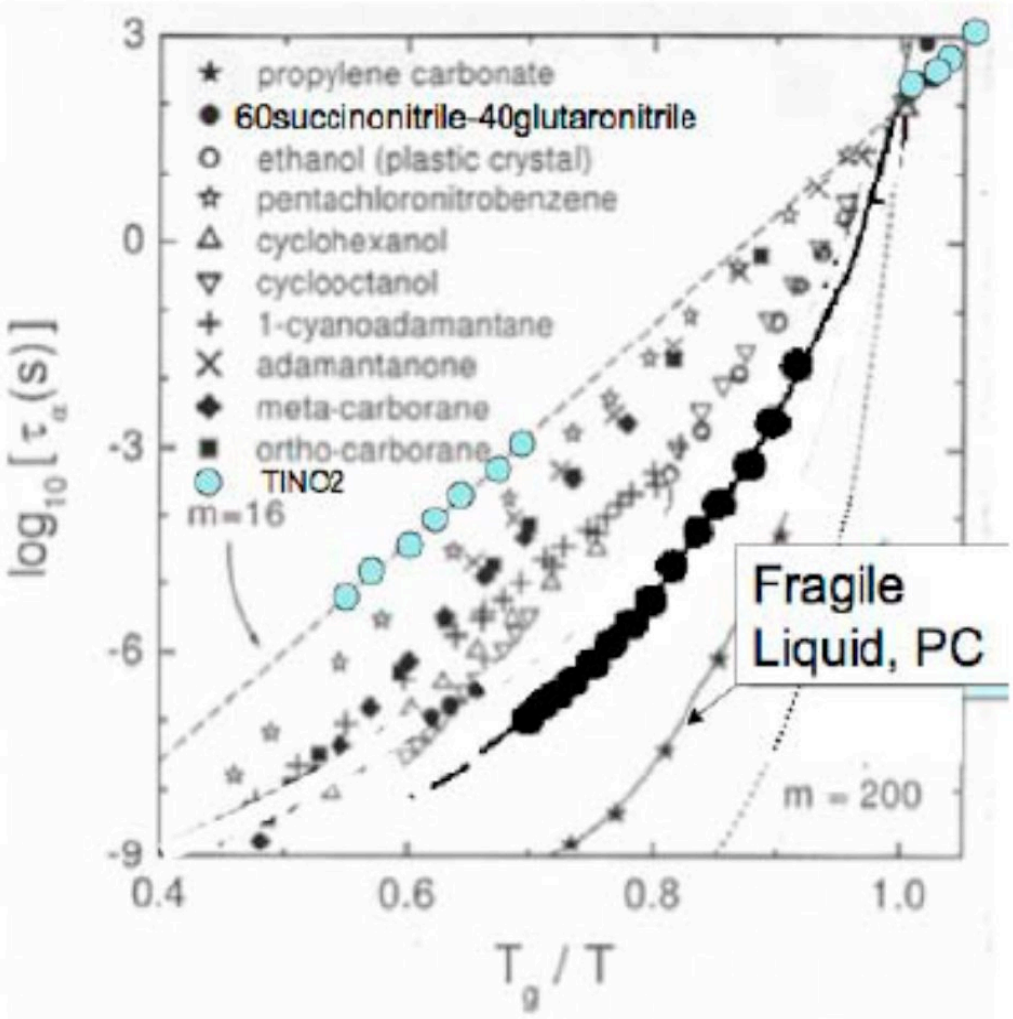
Rotator Glass, P_2S_3

Plastic crystal relaxation time master plot

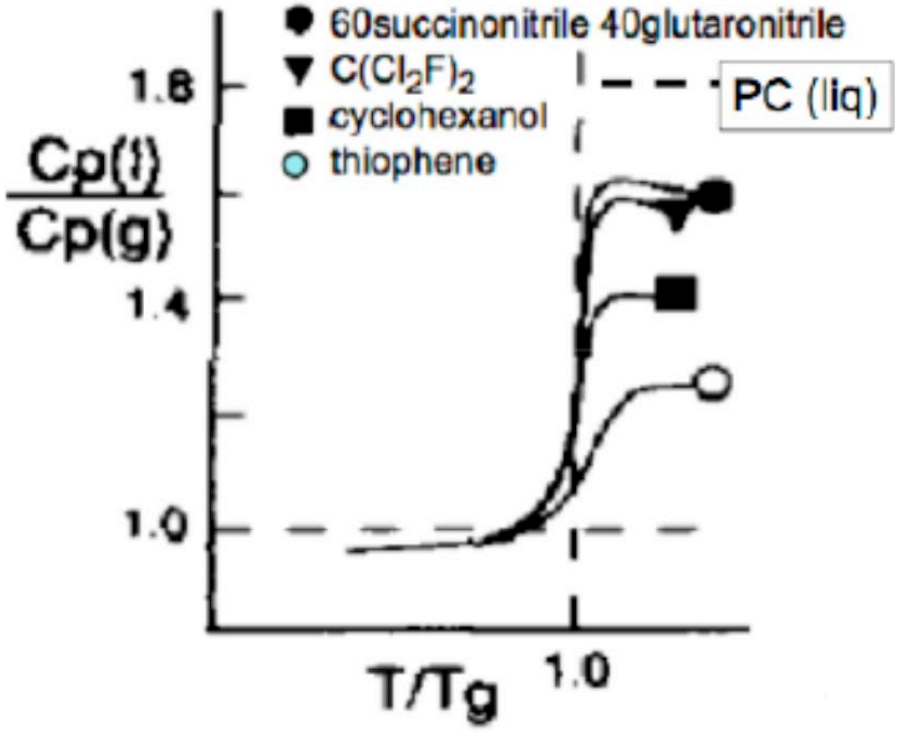


Suga in Japan, Bermejo, Ramos in Spain, Descamps in France, Nagel & Birge in the US, Bohmer Loidl in Germany

Plastic crystal rotational times and ΔC_p - look like viscous liquids, but more "strong". But same basic problems as for liquids.



But only rotational disorder involved



1,2 dimethylcyclohexane, from early Huffman et al study

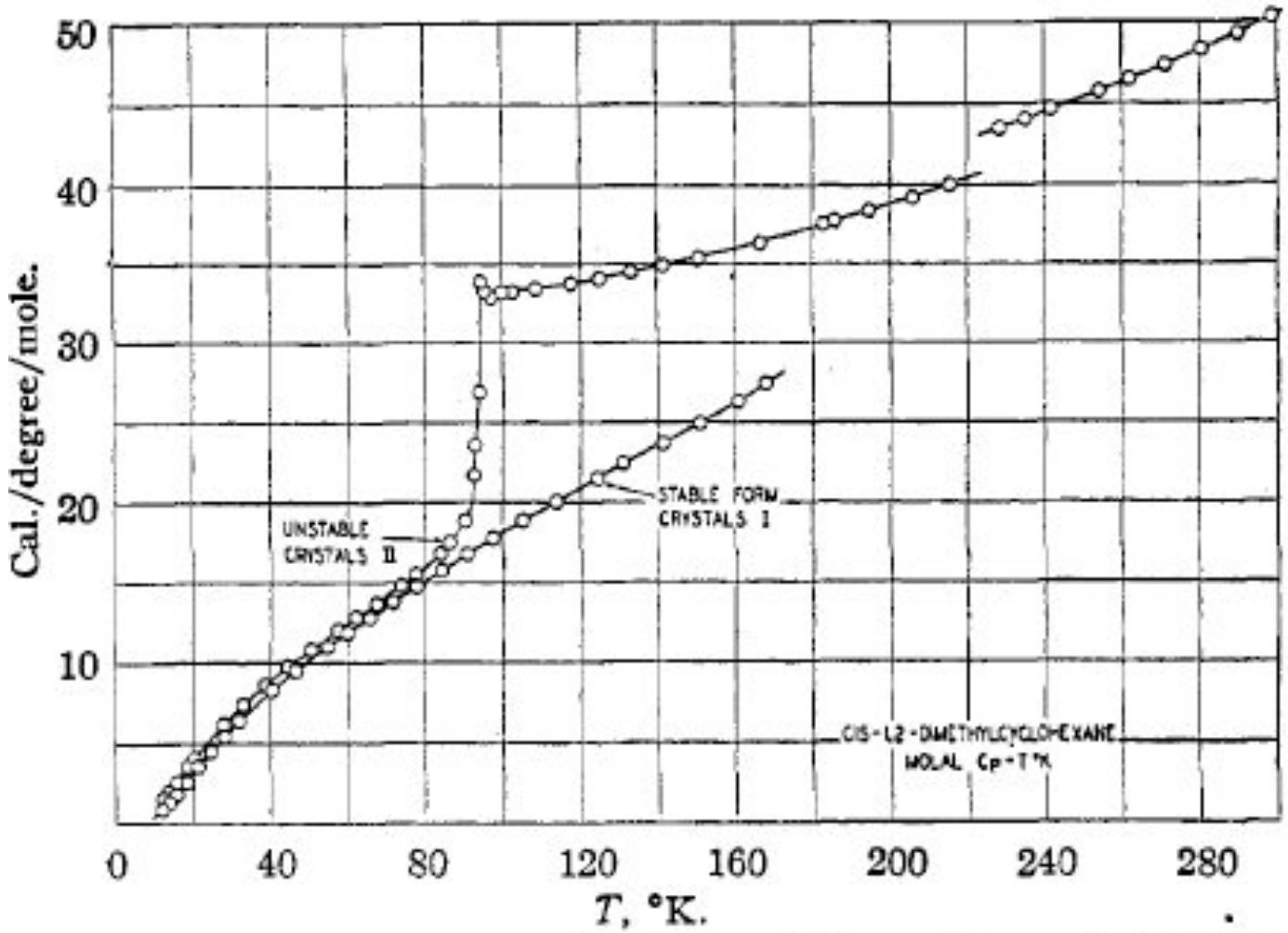


Fig. 2.—Heat capacity curves for the two different forms of *cis*-1,2-dimethylcyclohexane.

Work of Oguni and co-workers: adiabatic calorimetry enthalpy relaxation and dielectric relaxation in combination

Some of the least spectacular cases: non-fragile,

i.e like "strong" liquids, like SiO_2

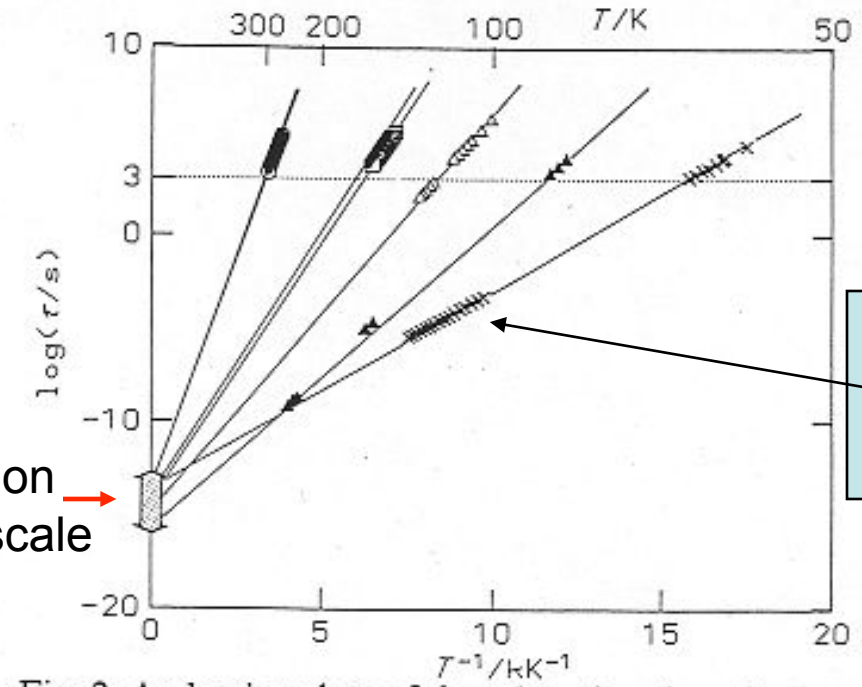
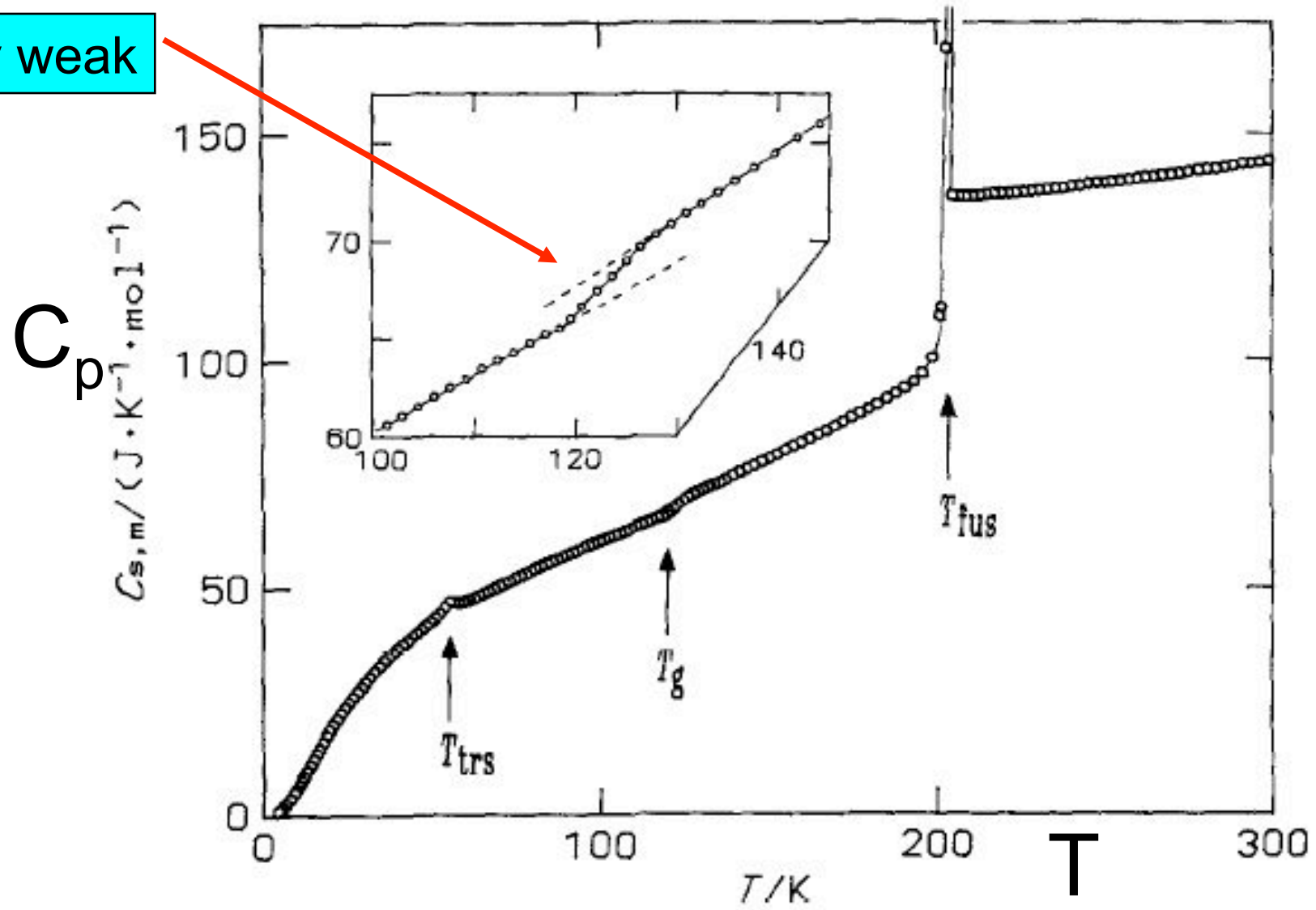


Fig. 2. Arrhenius plots of the relaxation times in some glassy crystals: ○, H_3BO_3 [17]; ●, D_3BO_3 [17]; □, $SnCl_2 \cdot 2H_2O$ [25]; ■, $SnCl_2 \cdot 2D_2O$ [25]; △, C_4H_3BrS [16]; ▲, C_{60} [26]; ×, $TlNO_3$ [27]. Since the activation processes in the stable crystalline phase do not ordinarily change with temperature, the relaxation times obey Arrhenius equation. Solid lines stand for the temperature dependencies of the relaxation times fitted by using the equation. The pre-exponential factors τ_0 are found to be in the range 10^{-13}–10^{-16} s which is reasonable from the frequencies usually observed for the molecular vibrations.

These also have a jump in heat capacity when the time scale crossing occurs

But awfully weak



When the melting doesn't occur first?

PHASE TRANSITION AND FREEZING OF IONIC DISORDER IN $CsNO_2$ AND $TlNO_2$ CRYSTALS†

KEIICHI MORIYA, TAKASUKE MATSUO and HIROSHI SUGA

J. Phys. Chem. Solids Vol. 44, No. 12, pp. 1103–1119, 1983

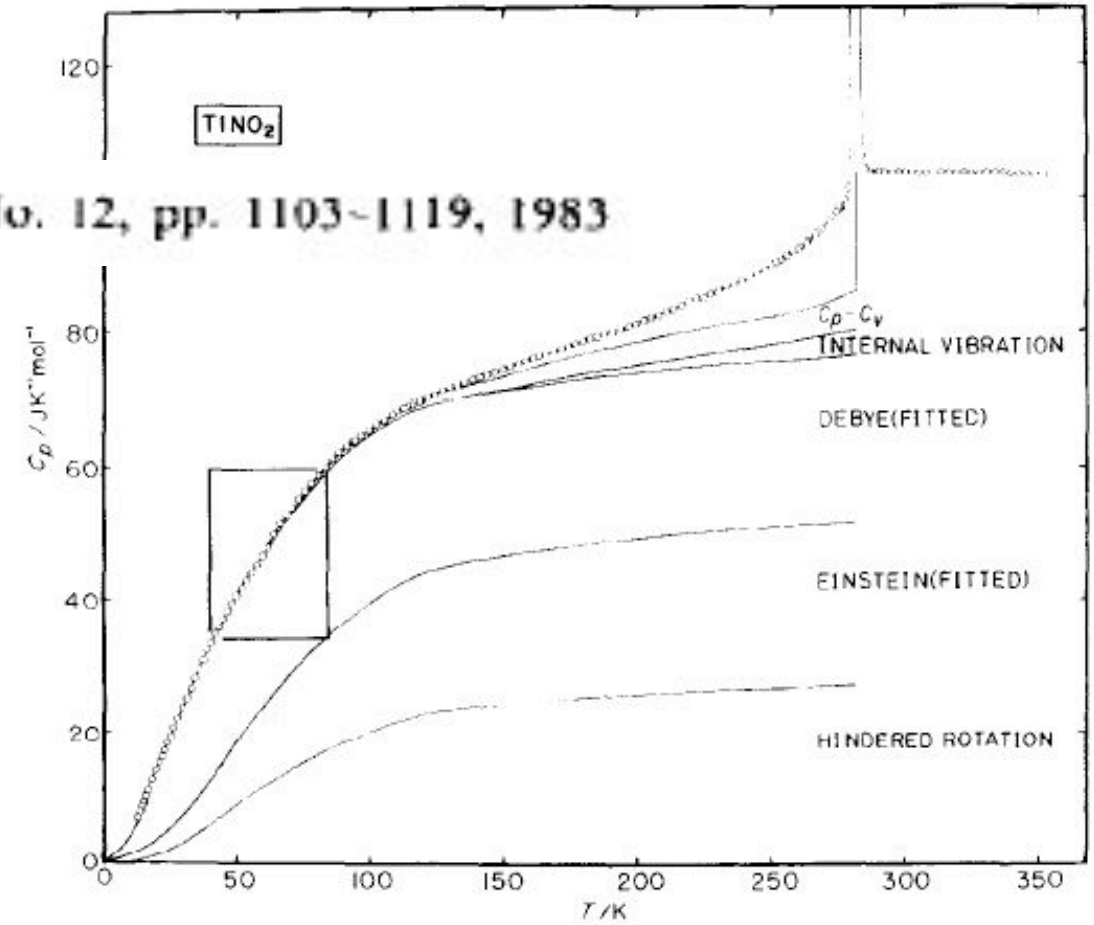


Fig. 2. The molar heat capacity of $TlNO_2$.

Lambda transition JUSSSST before melting

Smoking gun for the understanding of strong “glassy crystals” (they are the ones that melt before their lambda transitions)

So what happens in the cis-trans **mixed** crystal ?

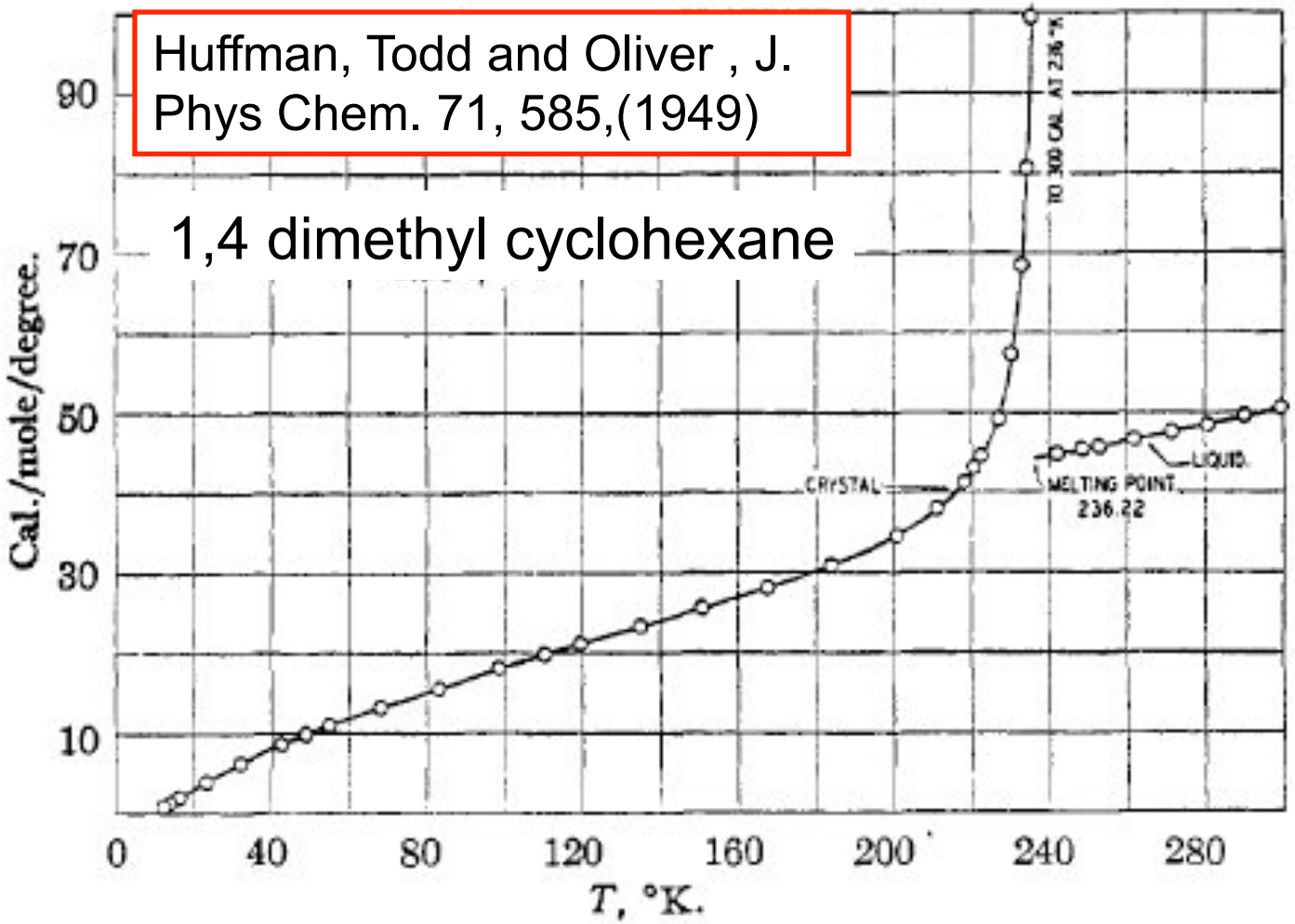
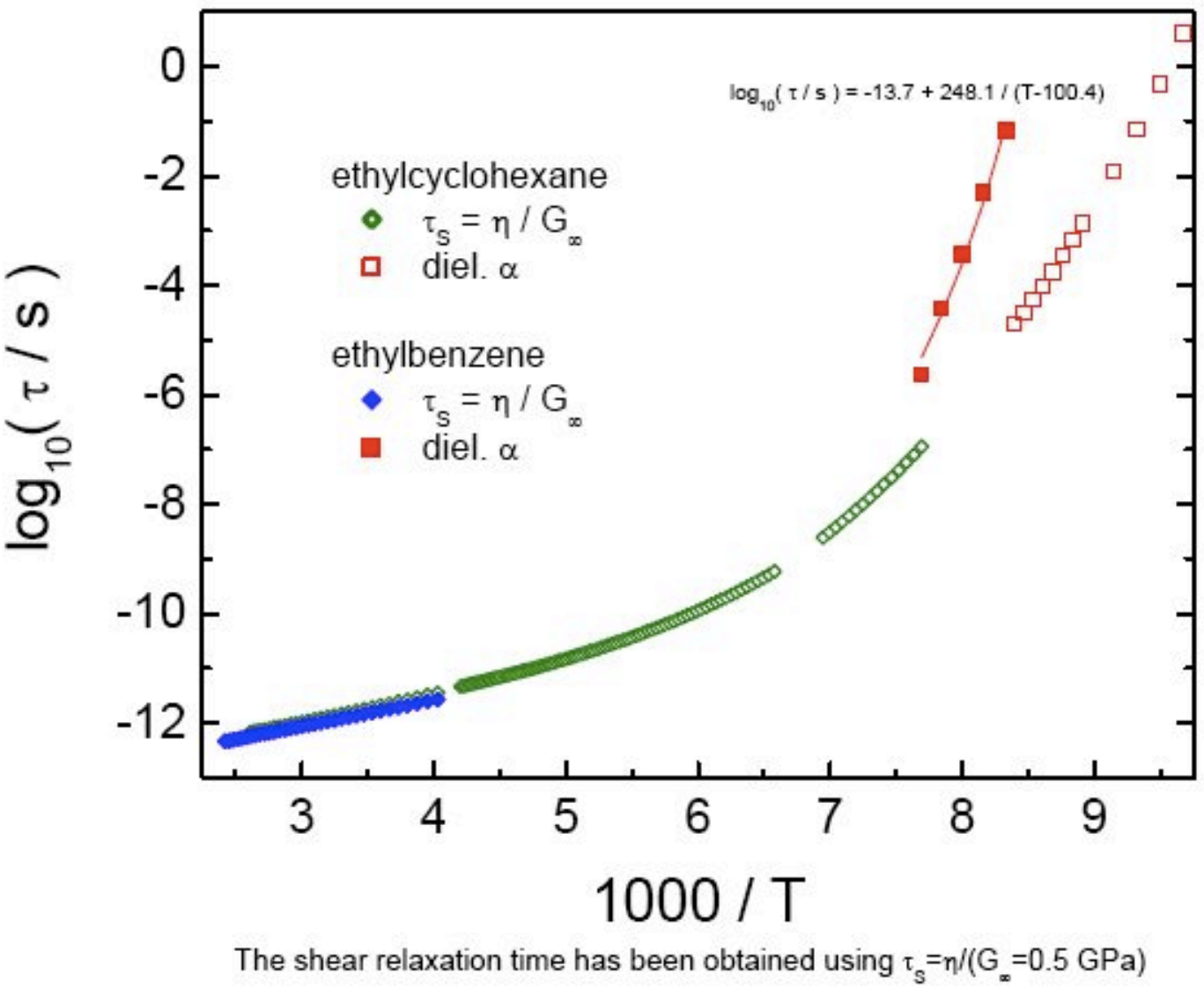


Fig. 3.—Heat capacity curve for *trans*-1,4-dimethylcyclohexane showing high heat capacity of crystals below the melting temperature.

BUT **ethylcyclohexane** is a glassformer !
though not fragile like ethyl benzene and toluene

(coming back to this, later)

Dielectric relaxation times for EB and ECH



The shear relaxation time has been obtained using $\tau_S = \eta/(G_\infty = 0.5\ \text{GPa})$

Lambda transition JUSSSST before melting

Smoking gun for the understanding of strong “glassy crystals” (they are the ones that melt before their lambda transitions)

Huffman, Todd and Oliver , J. Phys Chem. 71, 585,(1949)

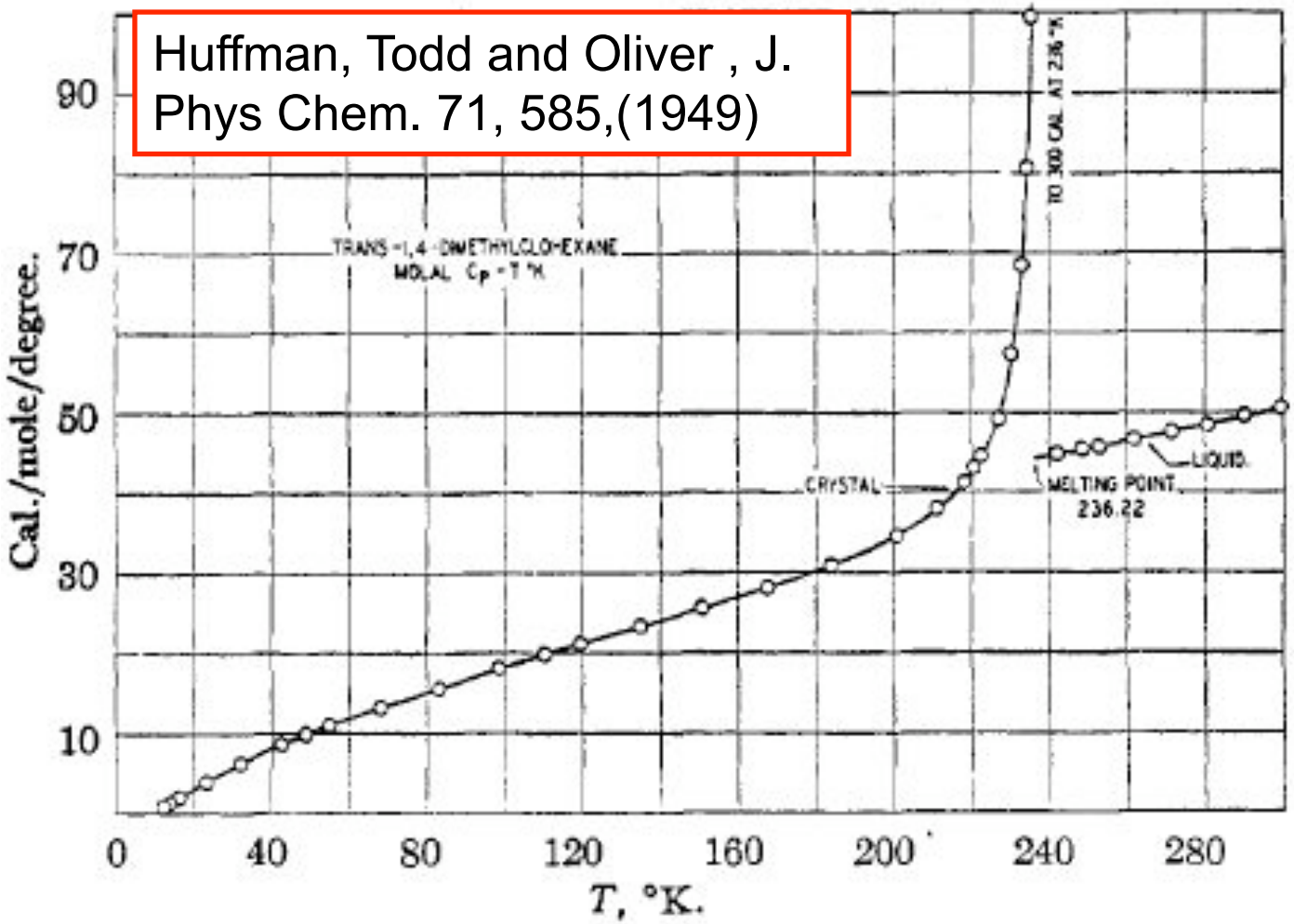
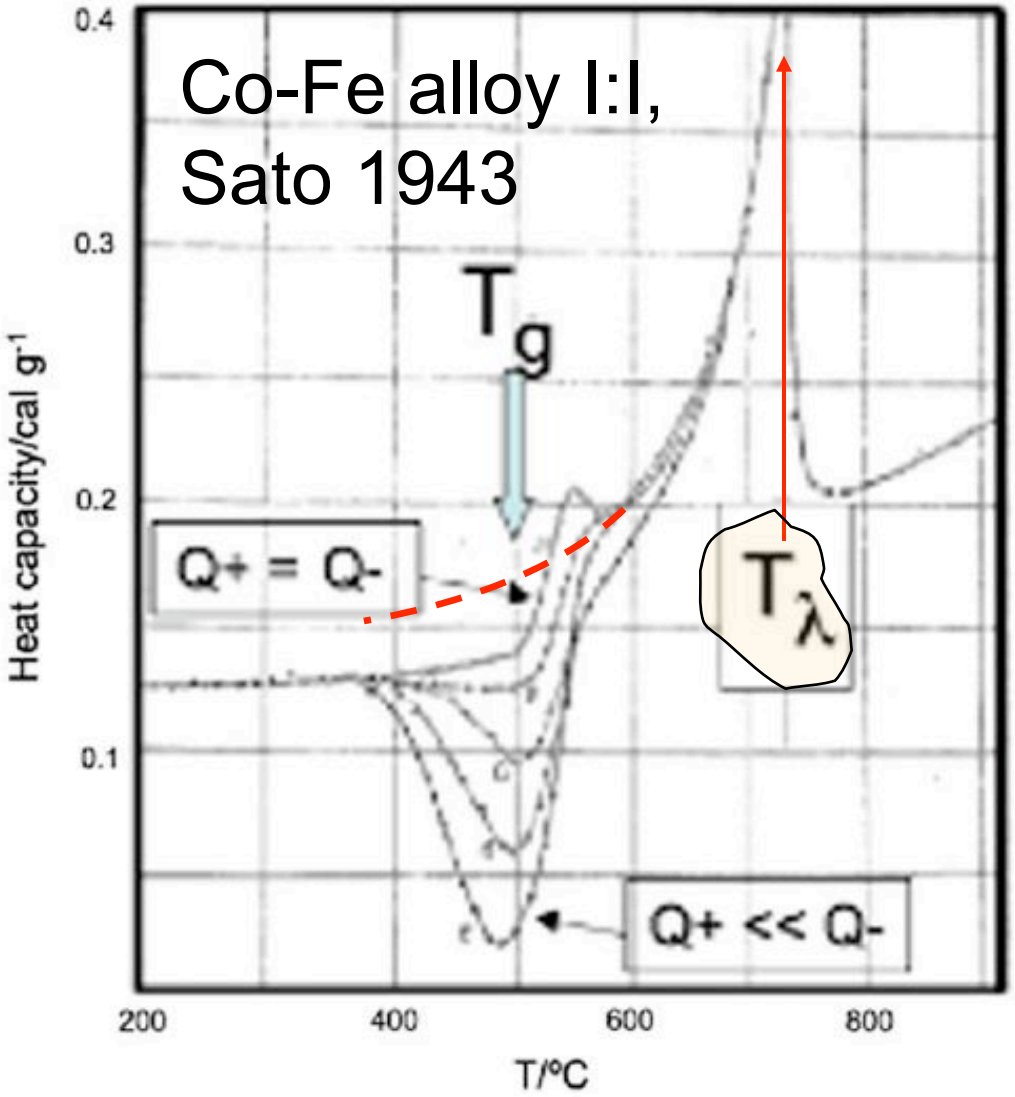


Fig. 3.—Heat capacity curve for *trans*-1,4-dimethylcyclohexane showing high heat capacity of crystals below the melting temperature.

Let's seek insights from a glass transition where background heat capacity is known

The kinetic arrest of ordering in a system with classical lambda transition heat capacity. The form is understood..There is no Kauzmann paradox here.

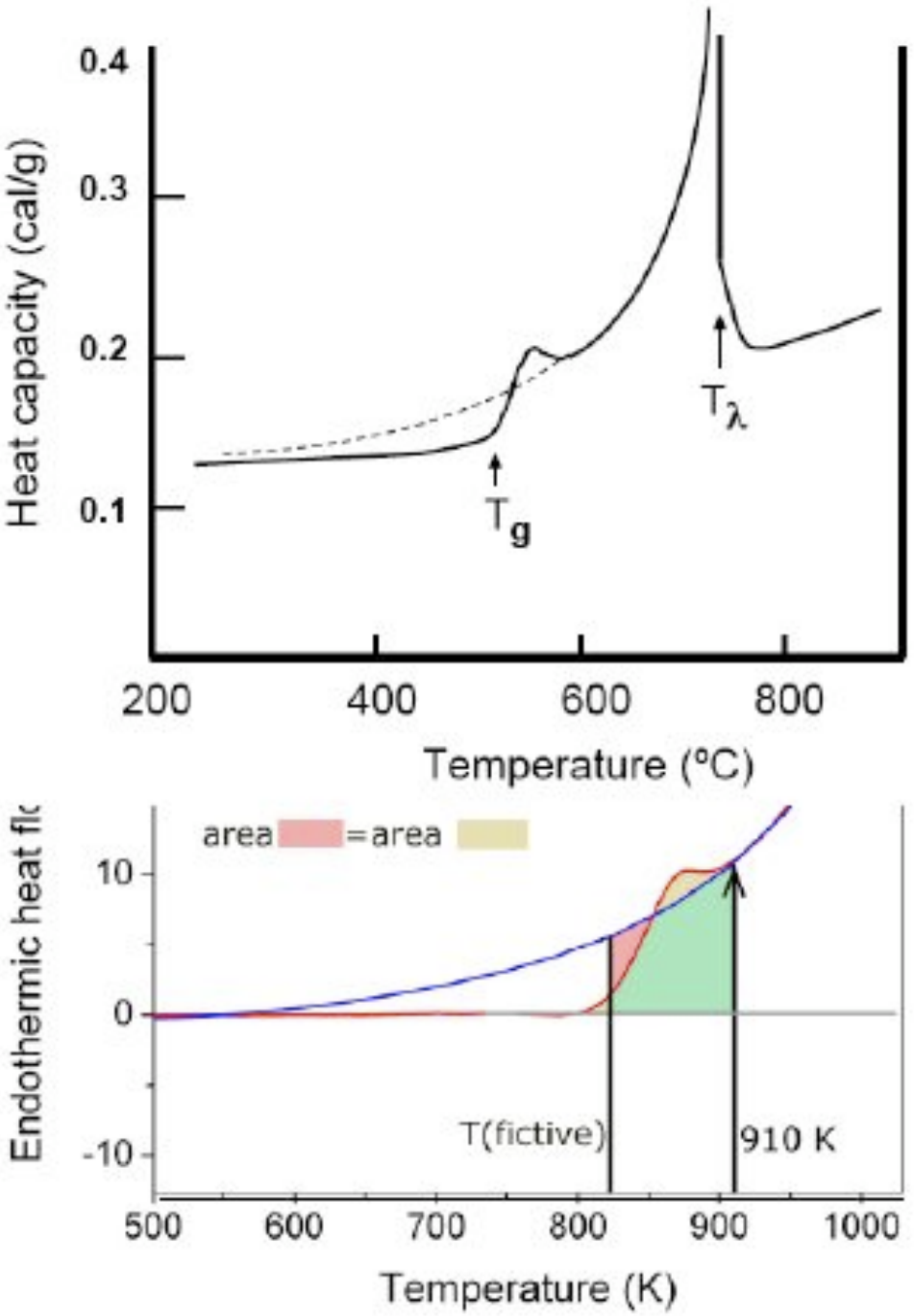
Effects of quenching followed by slow reheating are qualitatively as for normal glasses.



Glass transition in Co_{50}-Fe_{50} alloy.

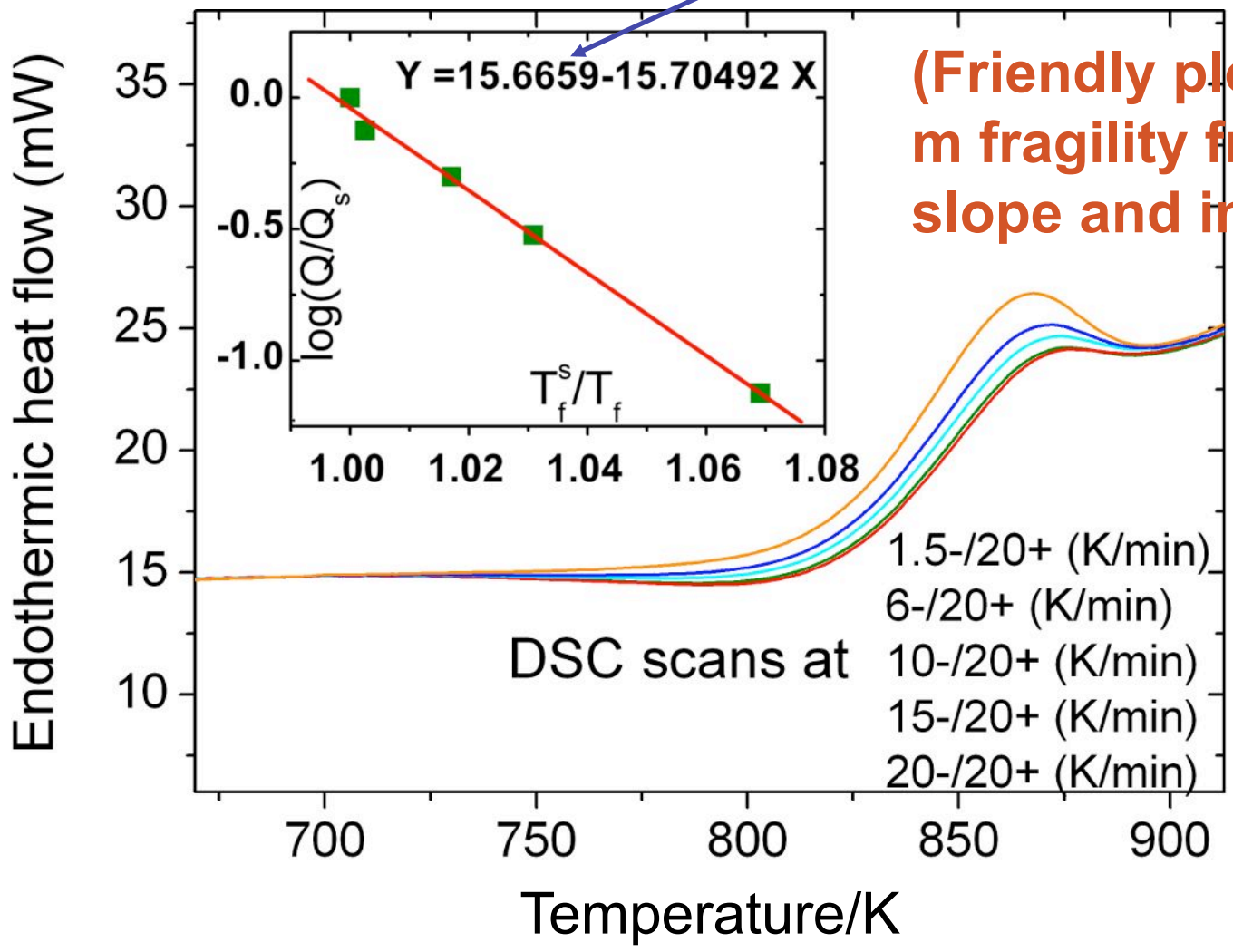
Collaboration with
Shuai Wei, Isabella Gallina and Ralf Busch

We study the ordering kinetics by determining the effect of cooling rate on the fictive temperature



Scans after cooling at different dT/dt, and assessment of T_f and m

Wang, Velikov, CAA: JCP

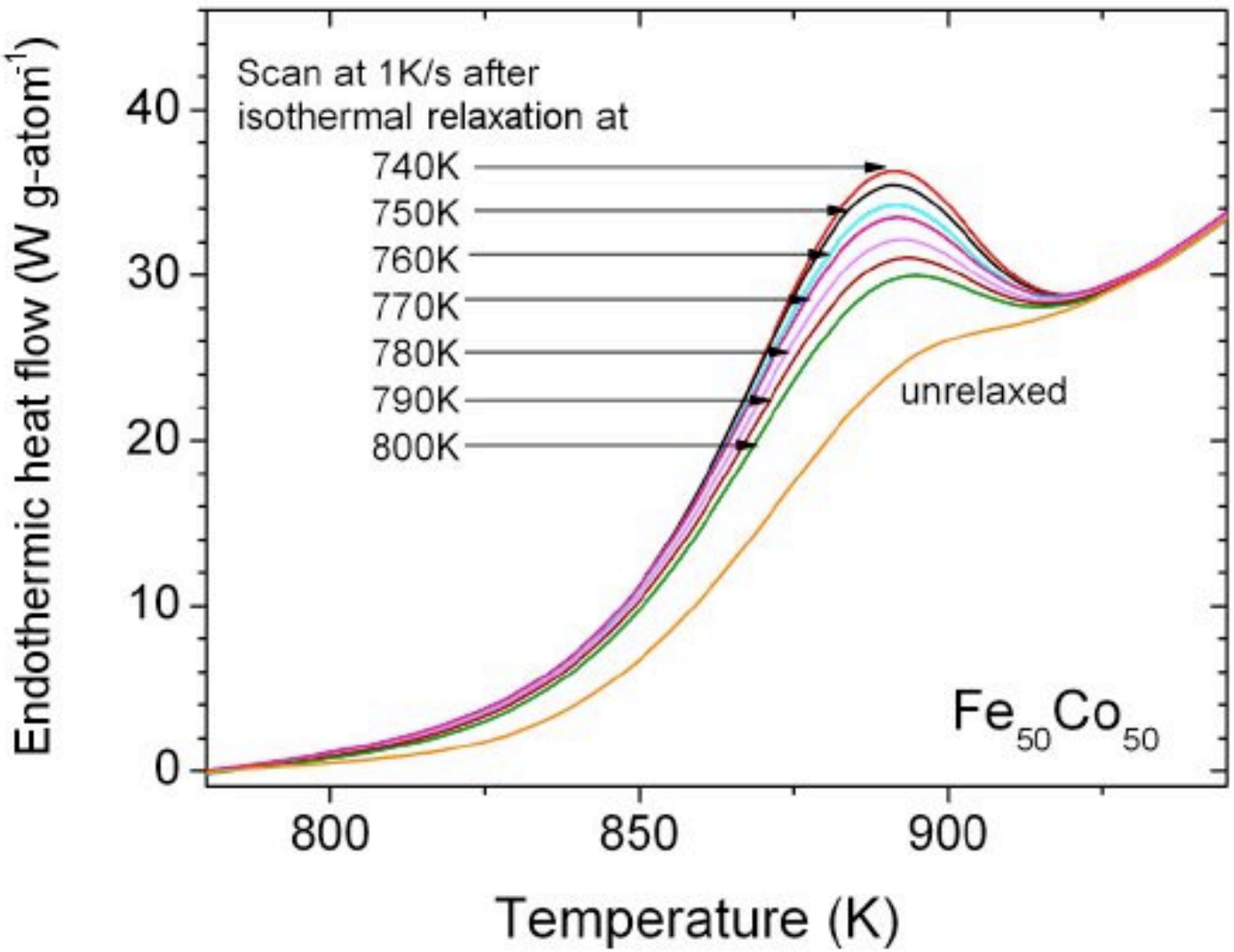


(Friendly plot gives m fragility from both slope and intercept)

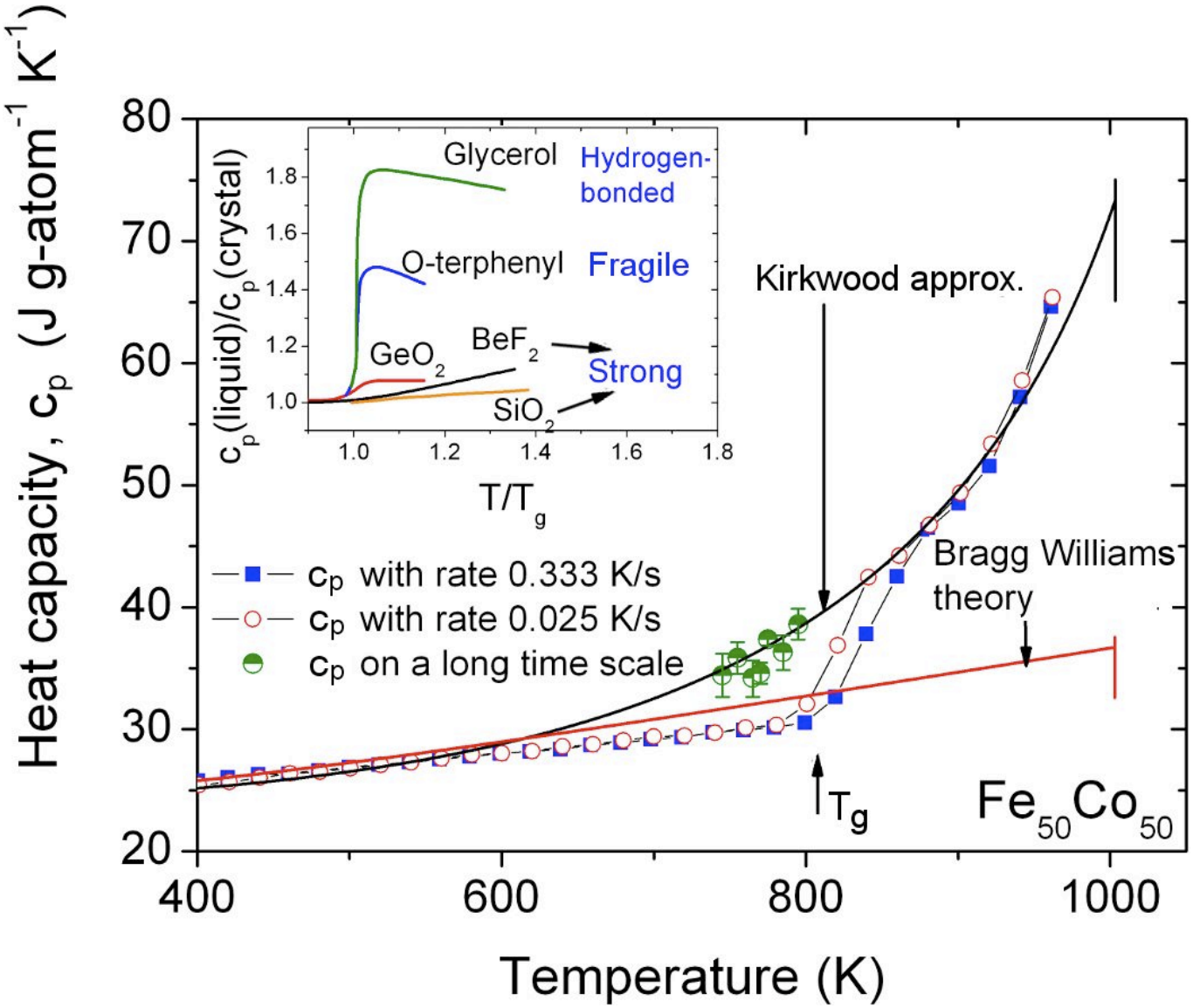
It’s Arrhenius !

And the slope is 16 like the ideal “strong” liquid

Enthalpy recovery after annealing at T



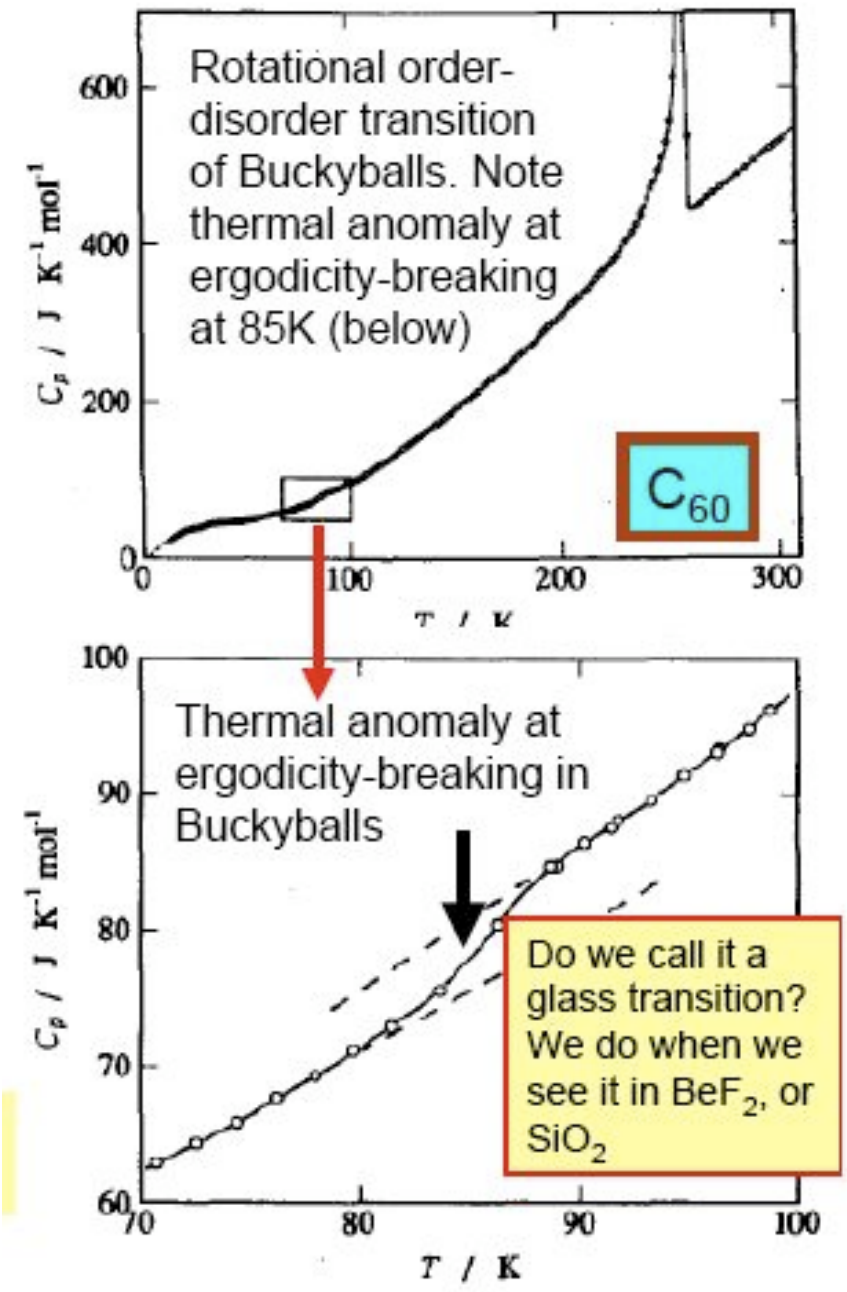
Equilibrium heat capacity in the Fe-Co superlattice below Tg



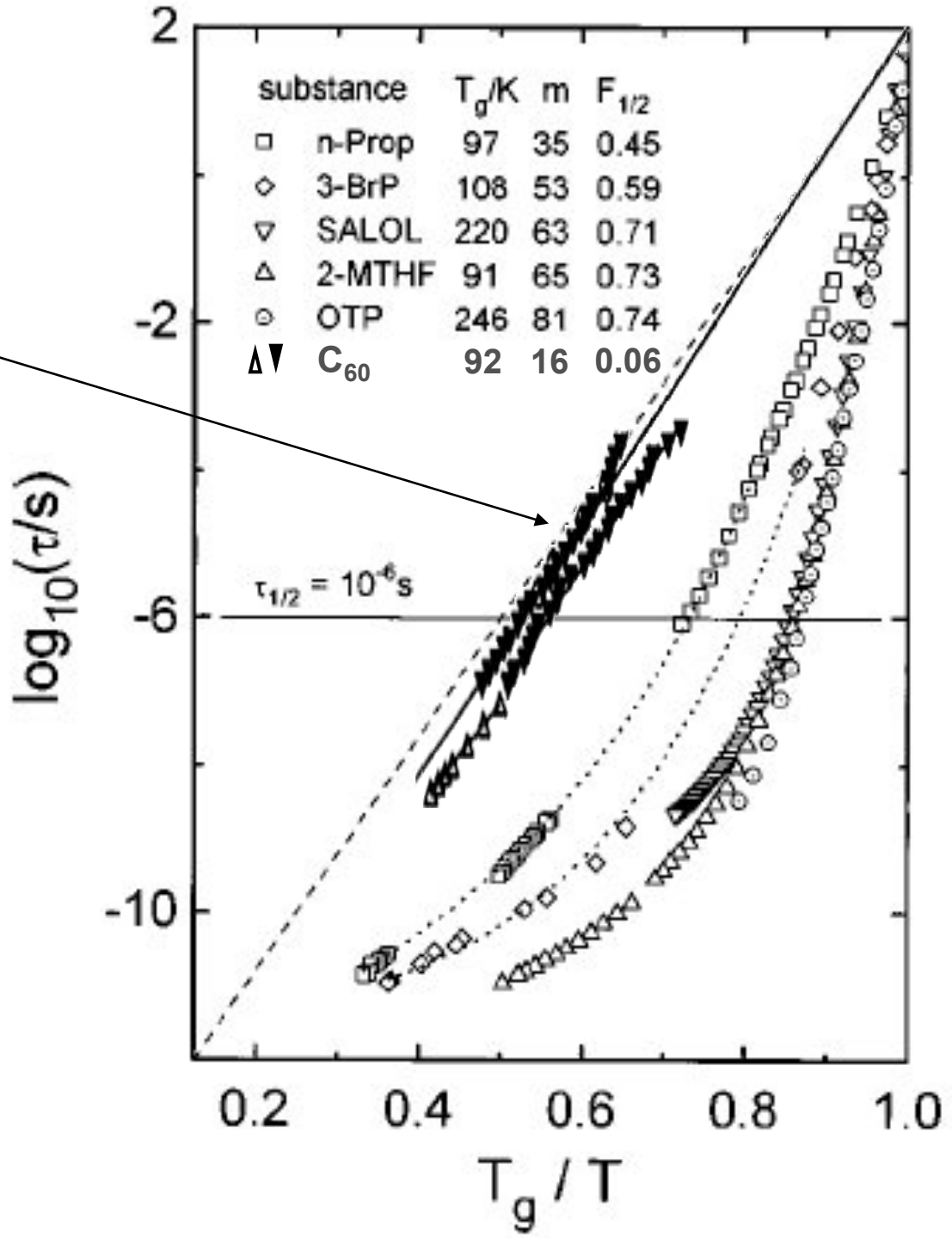
Important question:

Which way is the correlation length changing, as Tg is approached in ***THIS*** case?

Another glass transition at the foot of a lambda transition

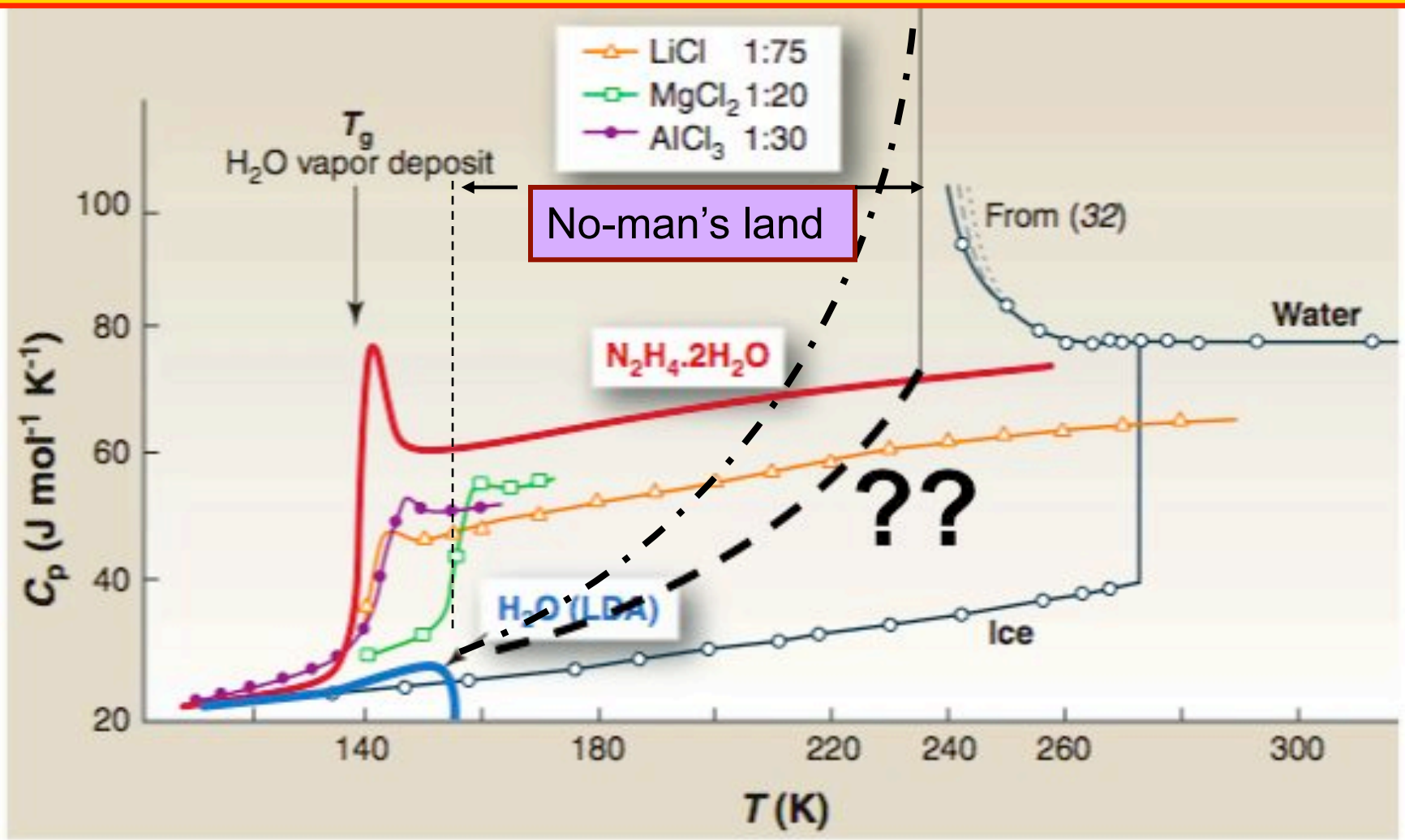


Dielectrics S&F with buckeyballs, relaxation is "strong", like SiO_2



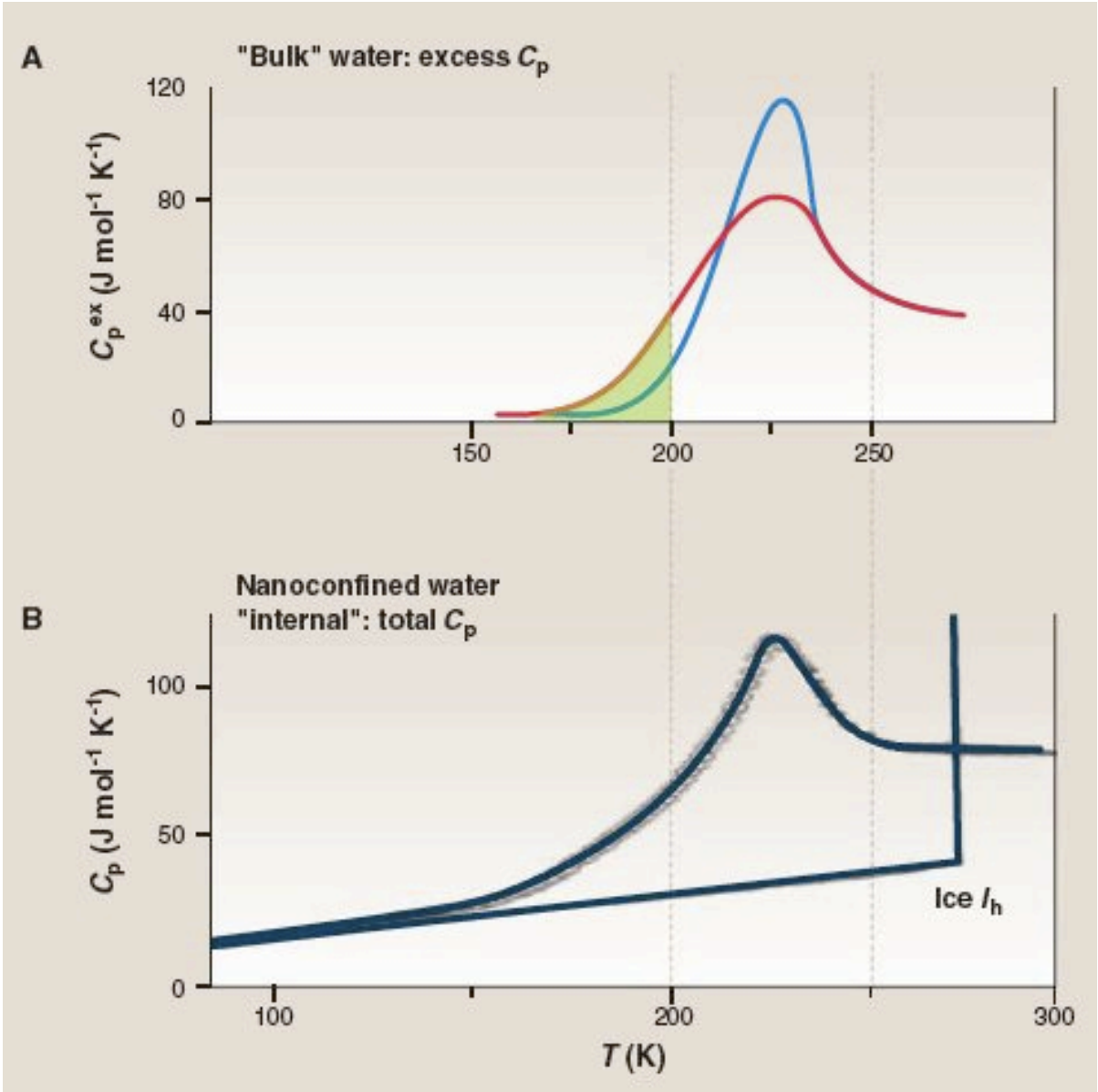
It reminds one of the vexing case of water - which I see as a Rosetta stone for understanding glassformers.

(Glassy water made by hyperquenching has an almost undetectible glass transition, especially relative to its solutions)

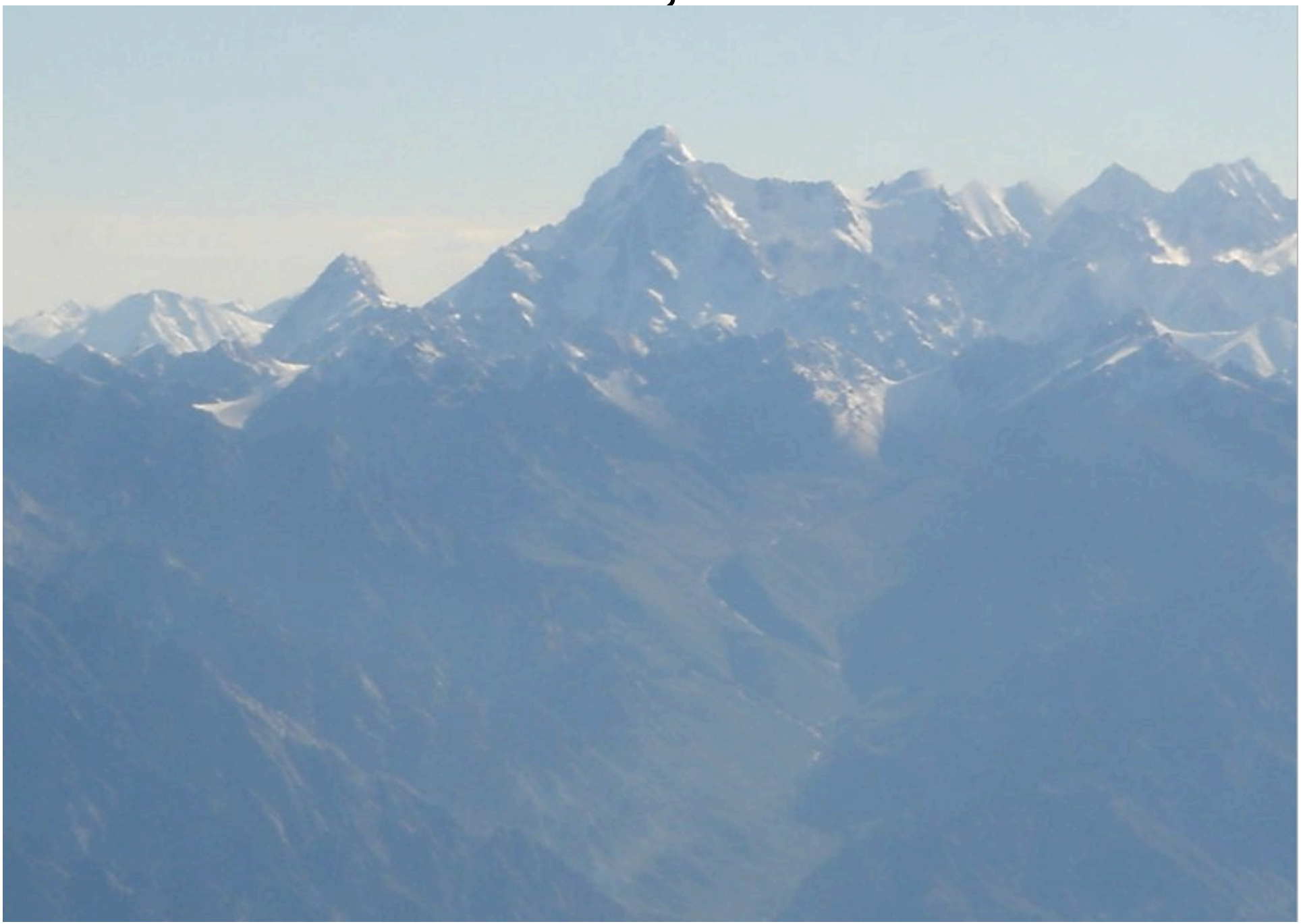


It reminds one of the vexing case of water - which I see as a Rosetta stone for understanding glassformers.

(from Science, 2008)



Tsien Shan Mtns, Western China



Outline for talk

A. BASIC CONCEPTS AND TERMS, and SYSTEMS

The glass transition for liquids- non-trivial thermodynamics

The energy landscape - and the rush to the top in some -Why the rush?

(fragile vs strong behavior)

B. HELP FROM NON-STANDARD CASES

Colloids, and crystals (rotator phases and disordering alloy superlattices) - and other λ transitions

C. LIQUID-LIQUID TRANSITIONS (visible and hidden) -

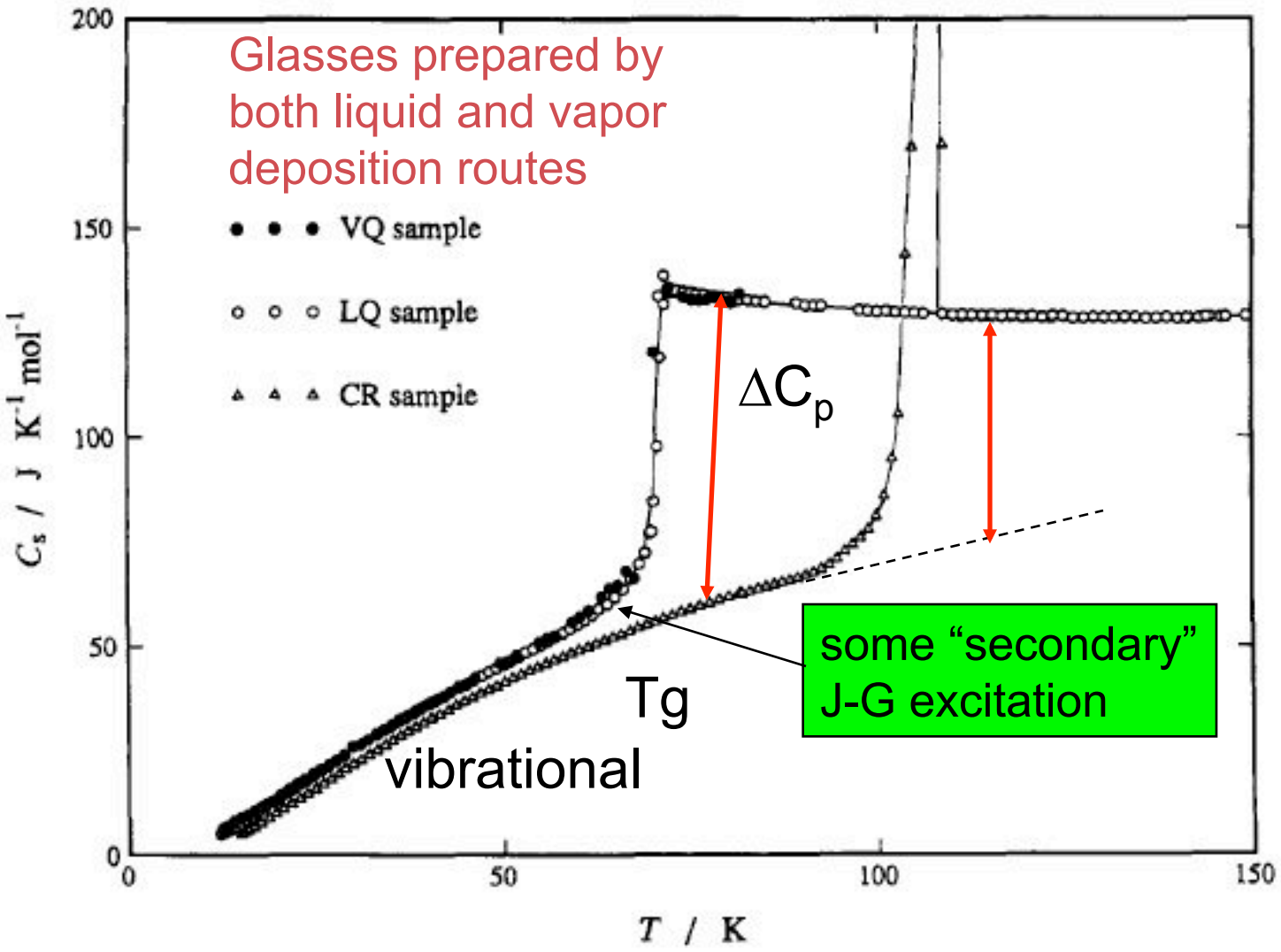
D. RELATION TO light scattering, neutron scattering, BMG phenomena, etc

OK: back to liquids to paint the BIG PICTURE

Heat capacity of 1 pentene, and the excess heat capacity behavior

Takeda et al (Osaka)

J. Phys. Chem., Vol. 99, N

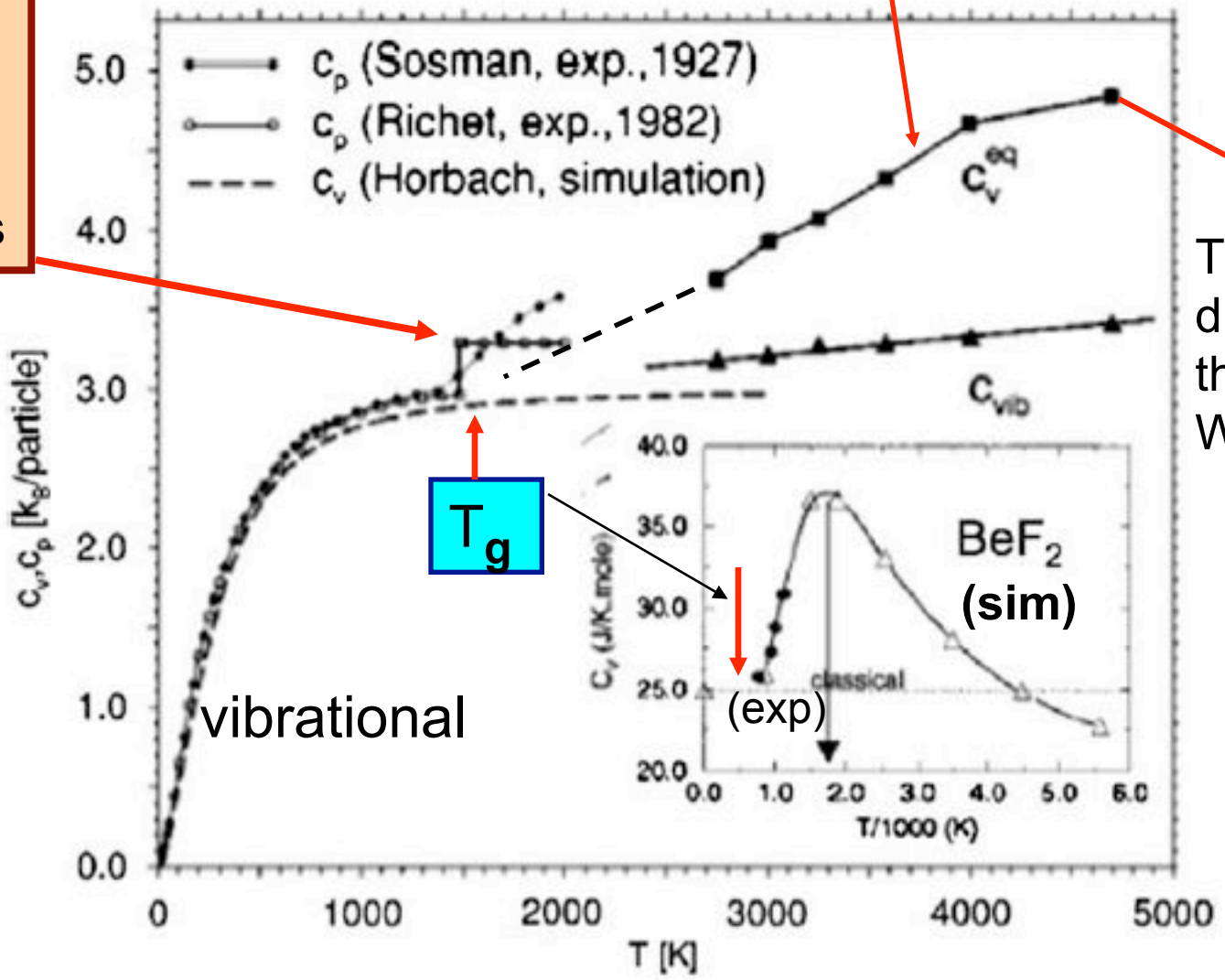


And silica at the other extreme

Horbach, Kob and Binder simulation "bone-dry" *(nothing in the lab so dry as a simulation with no water)*

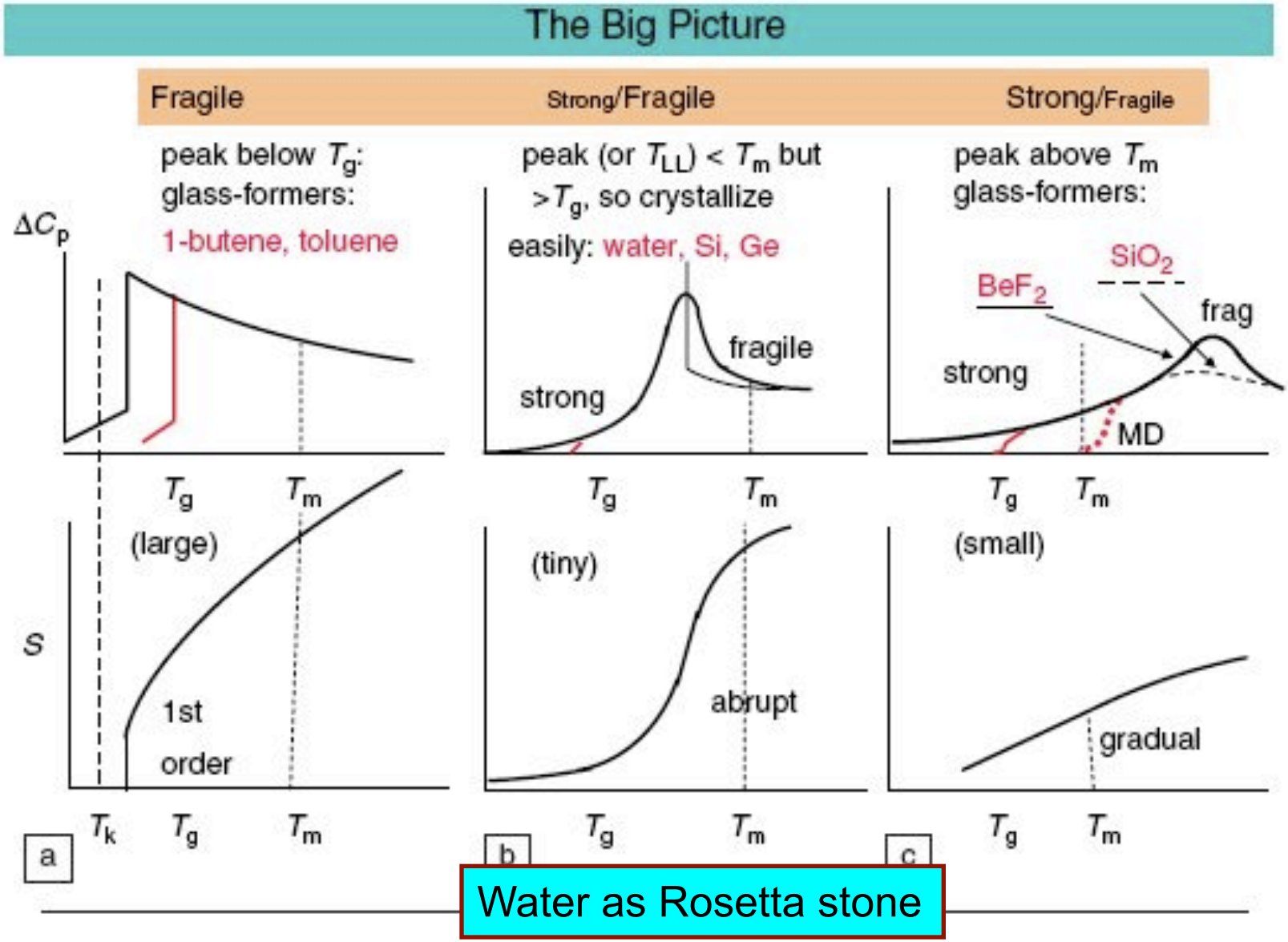
Tiny bit wet, means some broken

Si-O-Si bonds

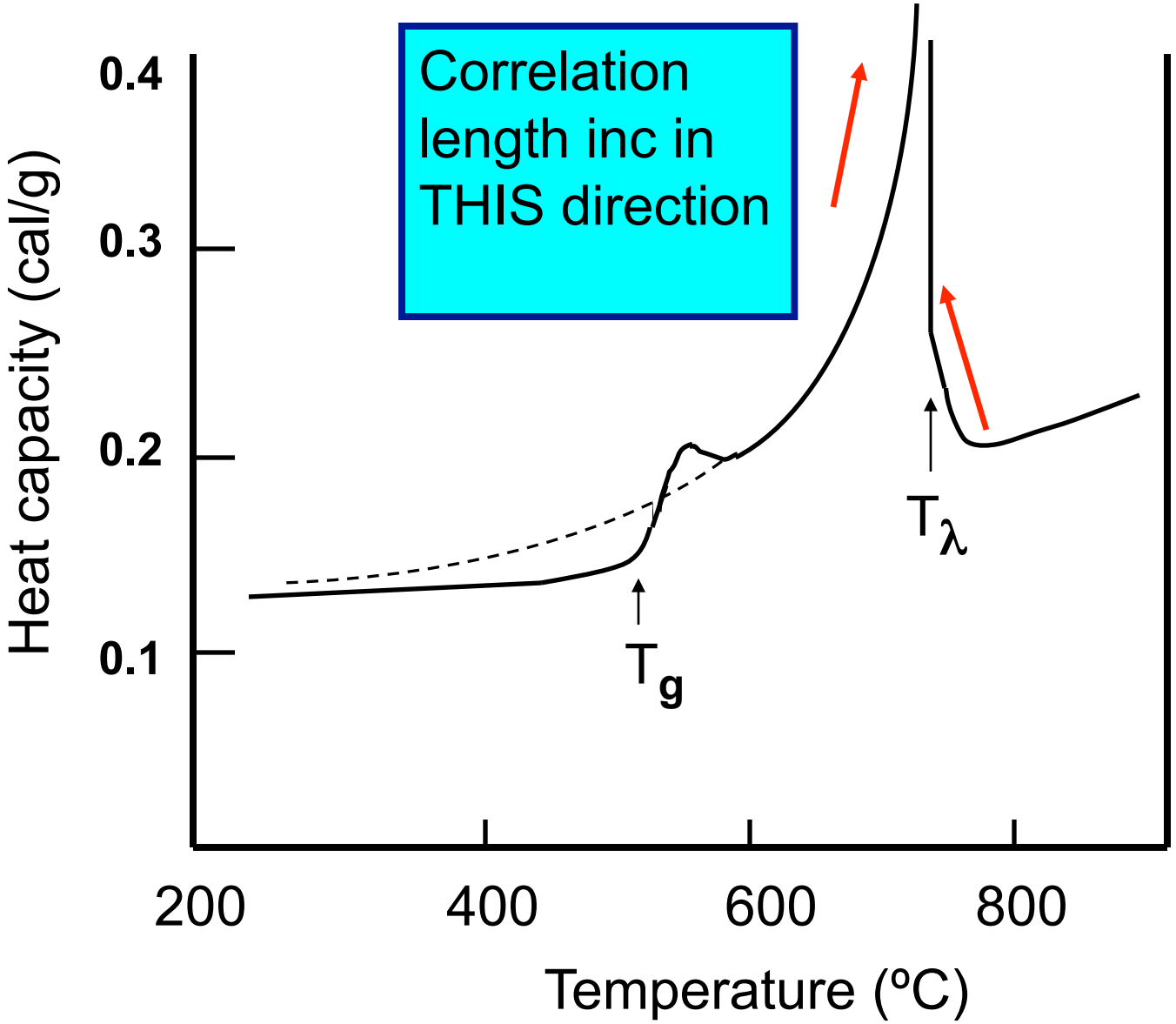


They didn't plot this point. Why?

Turnbull lecture, 2006 (MRS Bulletin, May 2008)

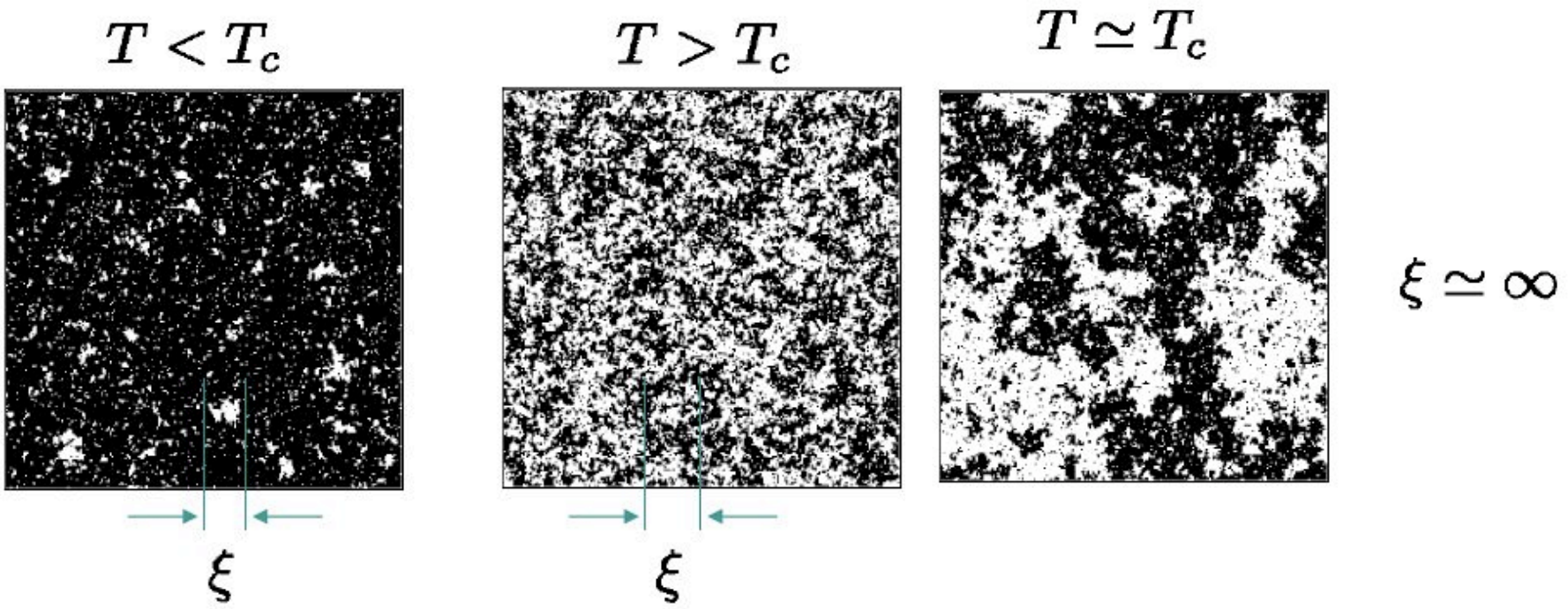


The Co:Fe system



Correlation lengths in the 2D Ising model

1. Correlation length



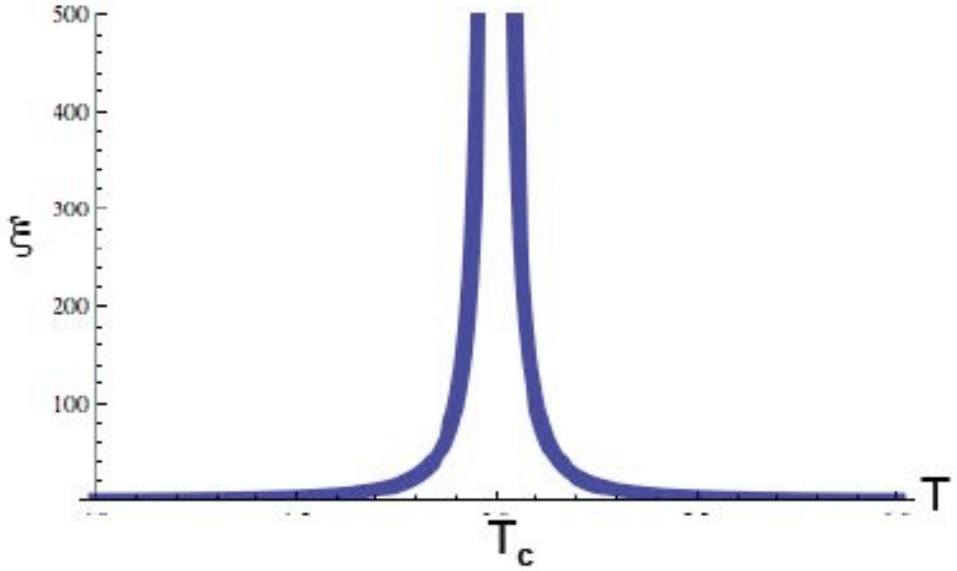
correlation length (in unit of lattice constant)

Correlation Length

At high T, the correlation length is low. The spins fluctuate rapidly, but they do so independently of each other.

Perhaps surprisingly, at low T, the correlation length is also low (even though the spins tend to be aligned). The point is that flipping σ_0 will hardly affect σ_n unless they are neighbors.

Near the critical temperature, the situation is different. The spins are constantly changing, but not independently. There are large domains of parallel spins which persist for a long time. Thus, spins far apart from each other are strongly correlated.



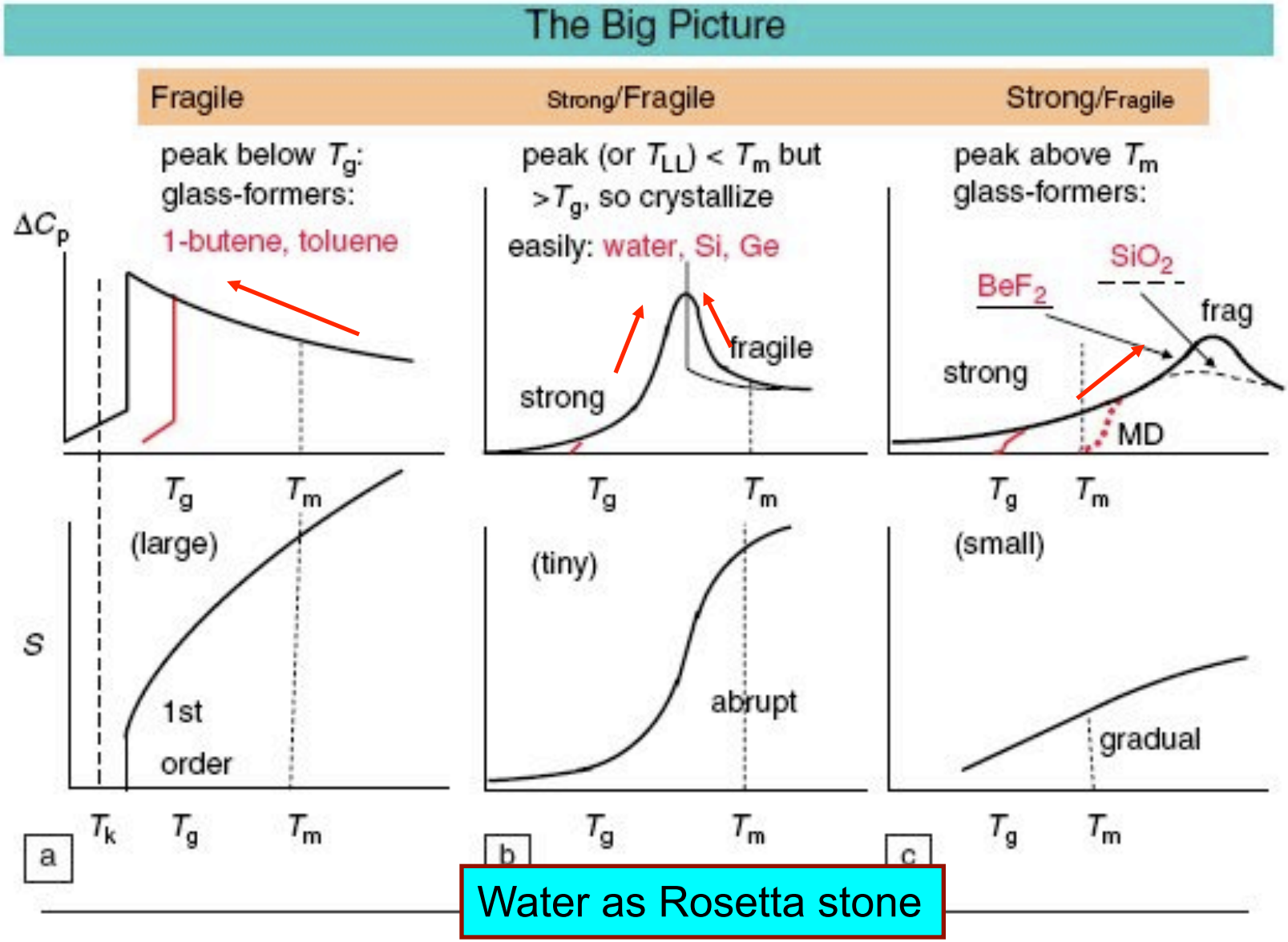
And now, a big picture, putting this all together with networks data

Glass-Formers and Viscous Liquid Slowdown since David Turnbull: Enduring Puzzles and New Twists

C.A. Angell

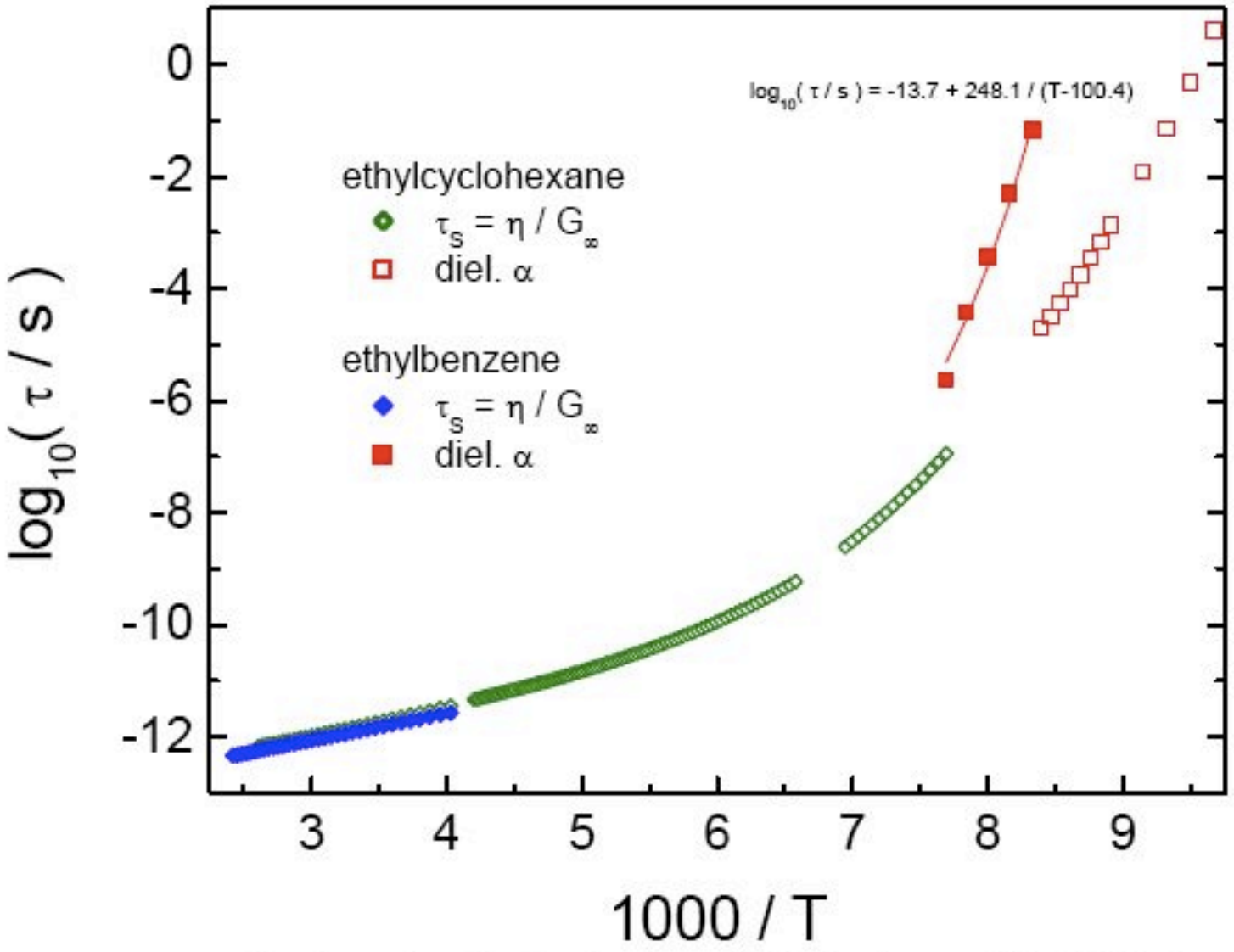
The following article is based on the 2006 Turnbull Lecture, presented by Austen Angell, on November 29, 2006, at the 2006 Materials Research Society Fall Meeting in Boston. The David Turnbull Lectureship is awarded annually by MRS to recognize the career of a scientist who has made outstanding contributions to understanding materials phenomena and properties through research, writing, and lecturing, as exemplified by David Turnbull of Harvard University.

Turnbull lecture, 2006 (MRS Bulletin, May 2008)



The Liquid-liquid transition below Tg

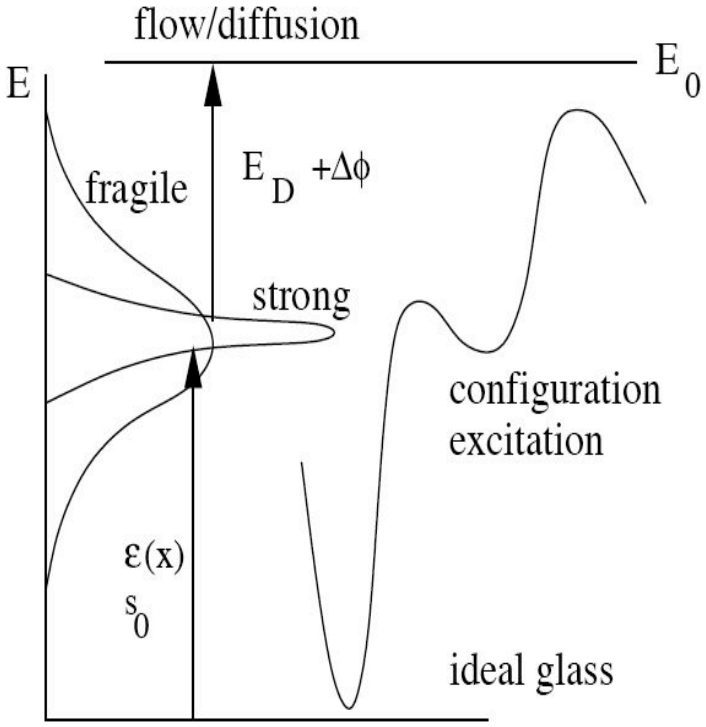
Dielectric relaxation times for EB and ECH

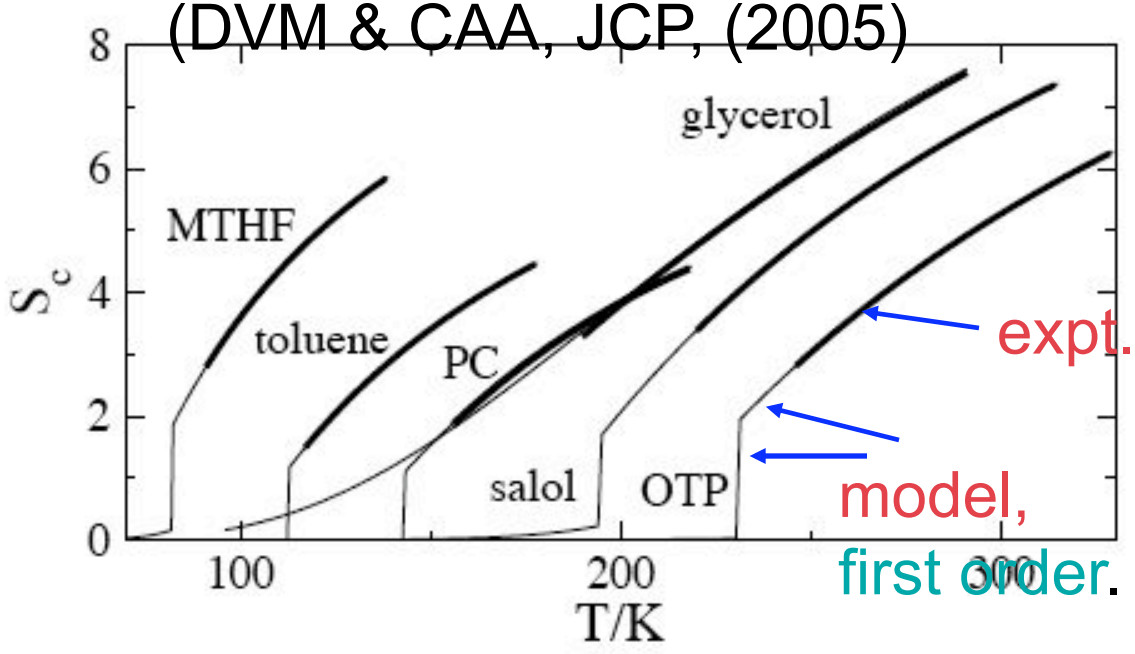


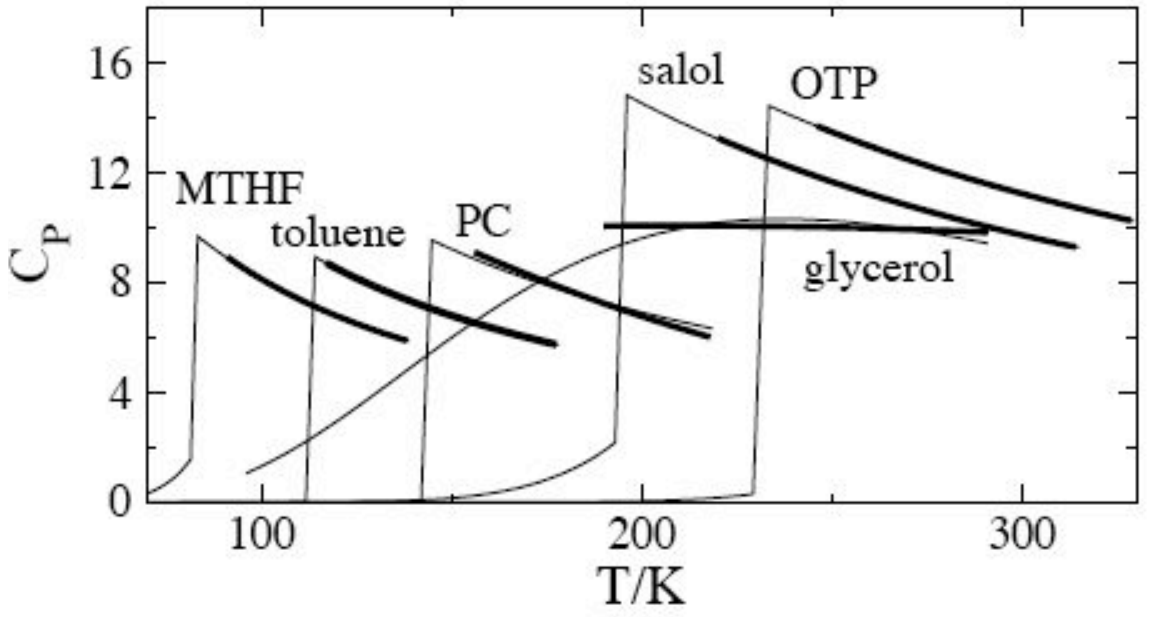
The shear relaxation time has been obtained using $\tau_S = \eta / (G_\infty = 0.5\ \text{GPa})$

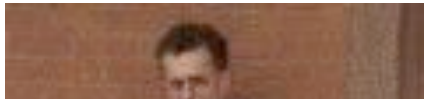
Dmitry Matyushov, ASU JCP 2007

(DVM & CAA, JCP, (2005)





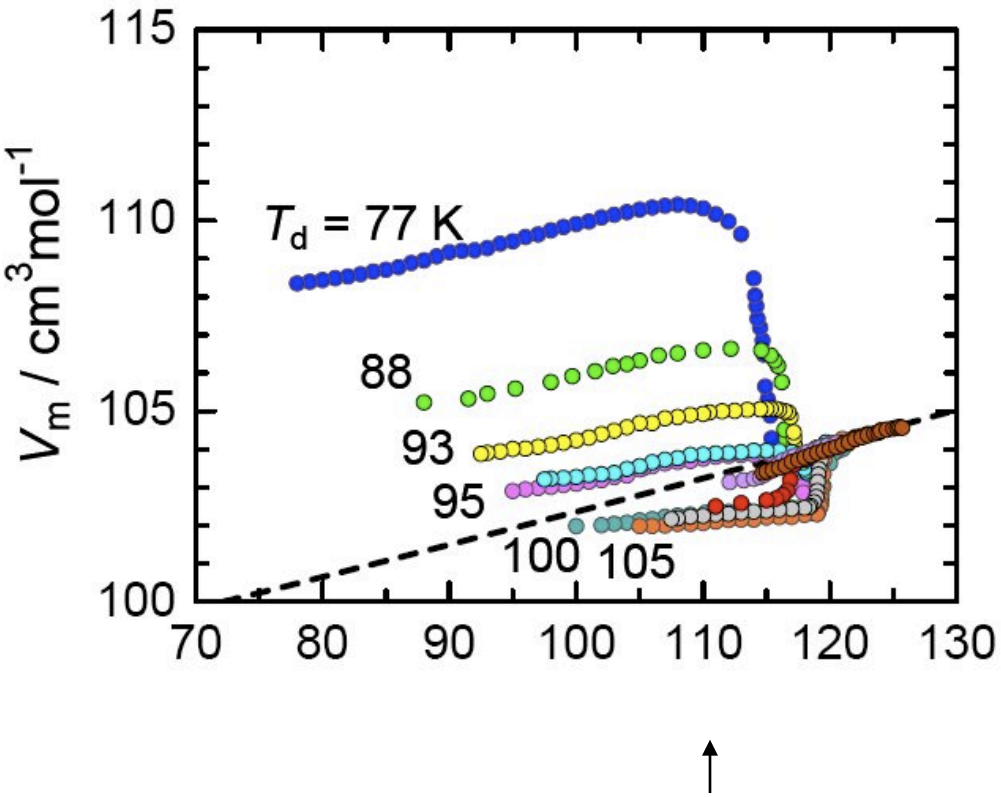




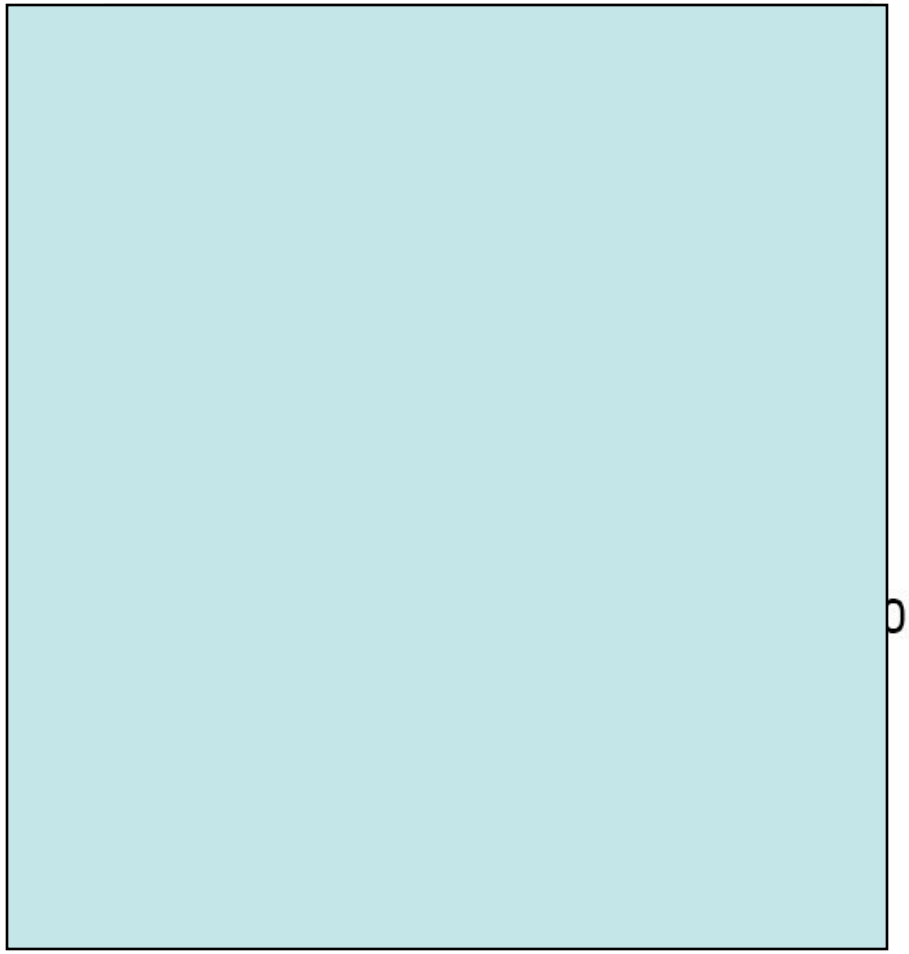
Vapor-deposited glasses (Fragile & Intermed.)

Ishii et al, Chem. Lett. (following Ediger and Co, “ultrastable glasses)

Ethylbenzene

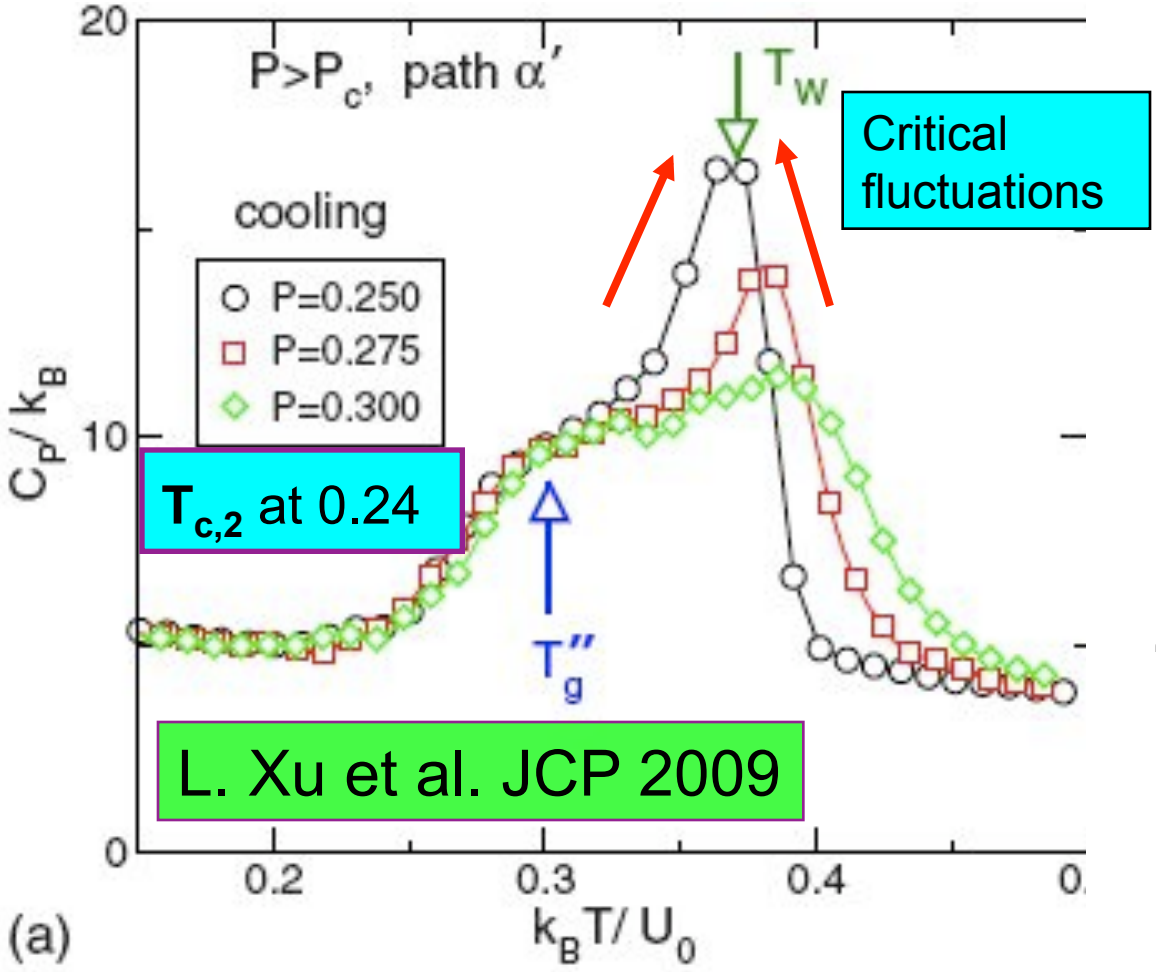


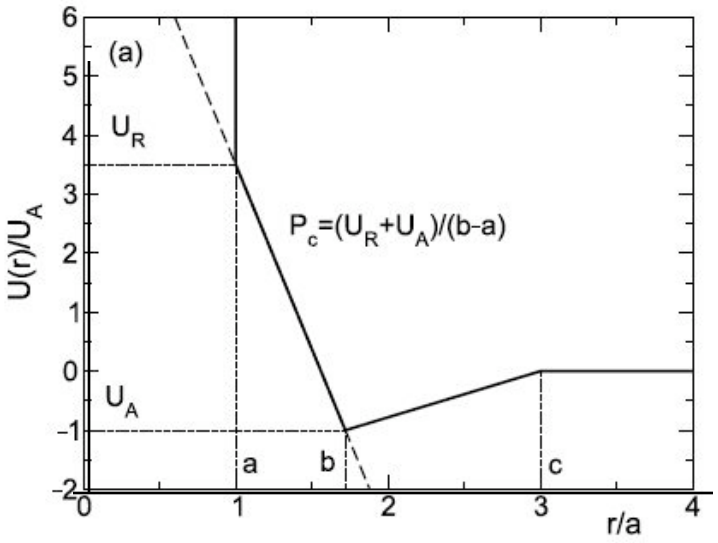
Ethylcyclohexane



T_g = 118, 122K
[SSA: JPC,82, 2627, (1978)

And here, finally, is a monatomic liquid doing the same thing - Jagla model





"attractive Jagla" model

Arxiv.1002.3206 chem.phys.

Glass transition with decreasing correlation length during cooling of $Fe_{50}Co_{50}$ superlattice and strong liquids.

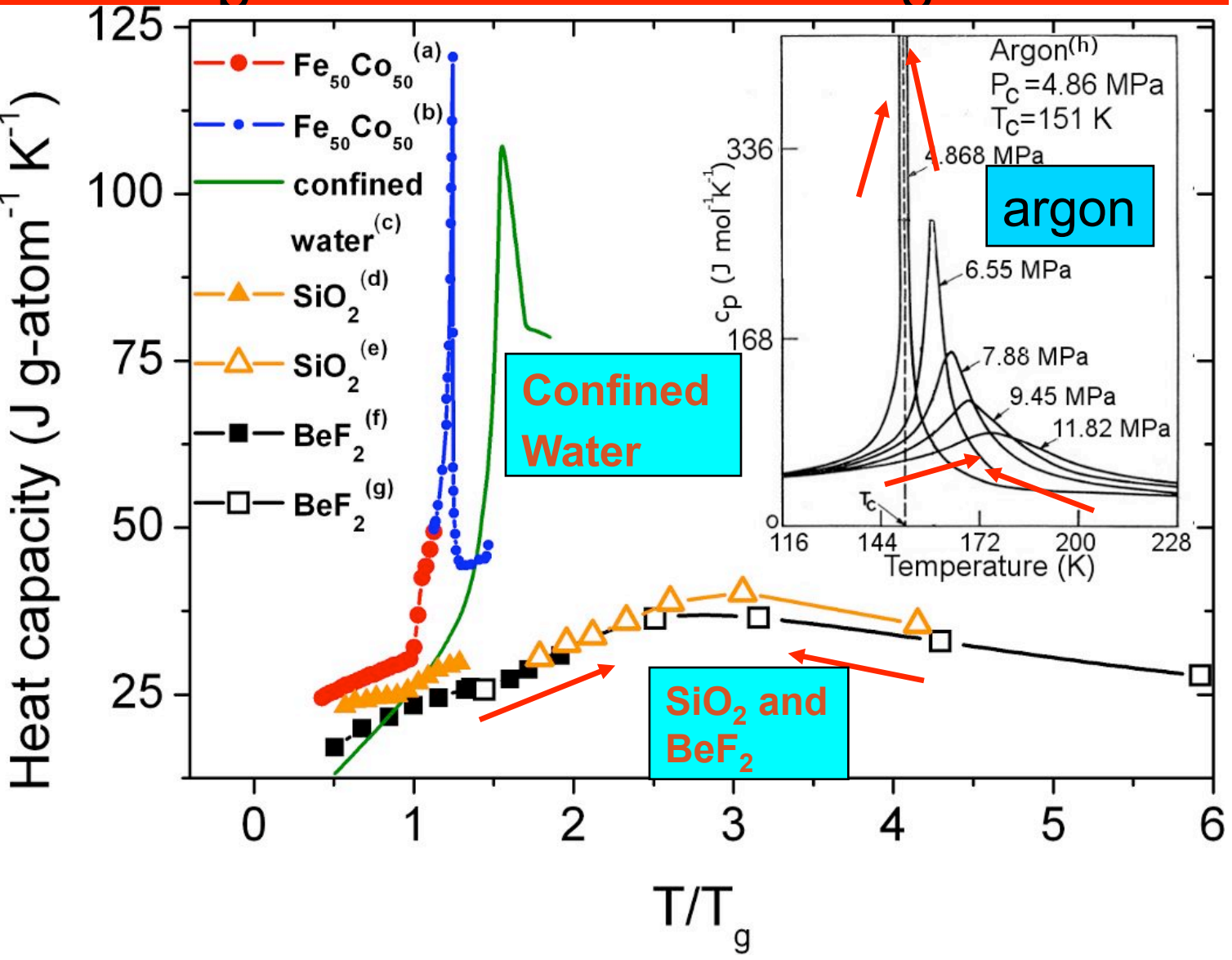
Shuai Wei@, Isabella Gallino@, Ralf Busch@ & C. Austen Angell*

@ Materials Science and Engineering Department, Saarland University, Universität Campus, 66123 Saarbruecken, Germany

*Department of Chemistry and Biochemistry, Arizona State University, Tempe, AZ 85287,
USA

Comparison of liquid C_p behavior with C_p near the L-G T_c

From Shuai Wei et al, (under review with Nature Physics)



SiO_2 P-V curve flattening out

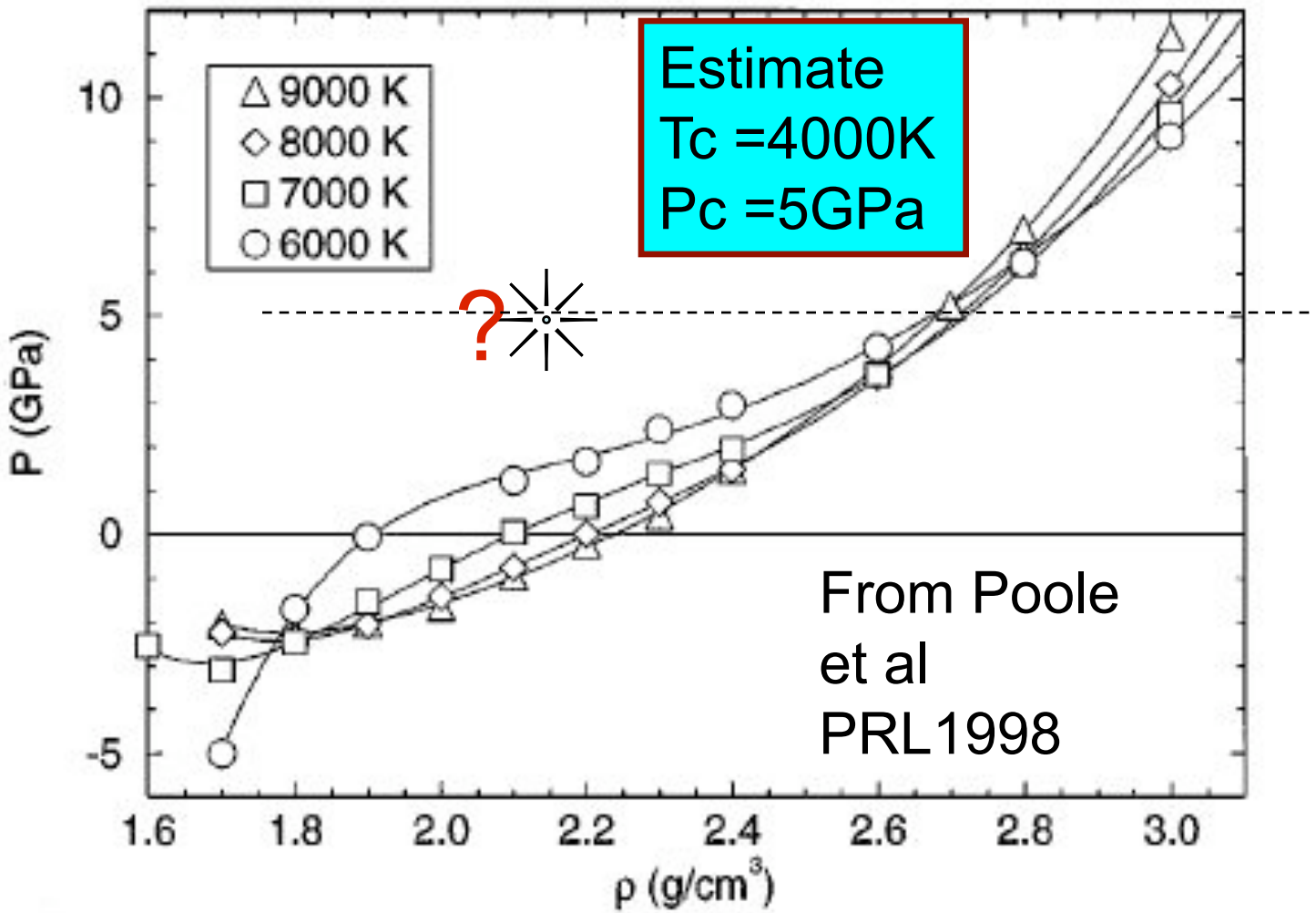


FIG. 3. Isotherms of P as a function of ρ. The solid lines are fits of the data to a sixth-order polynomial.

WAC SiO_2 (Poole-Hemmati) compress-ibility

Maximum coming up at density of 2.15 g/ml

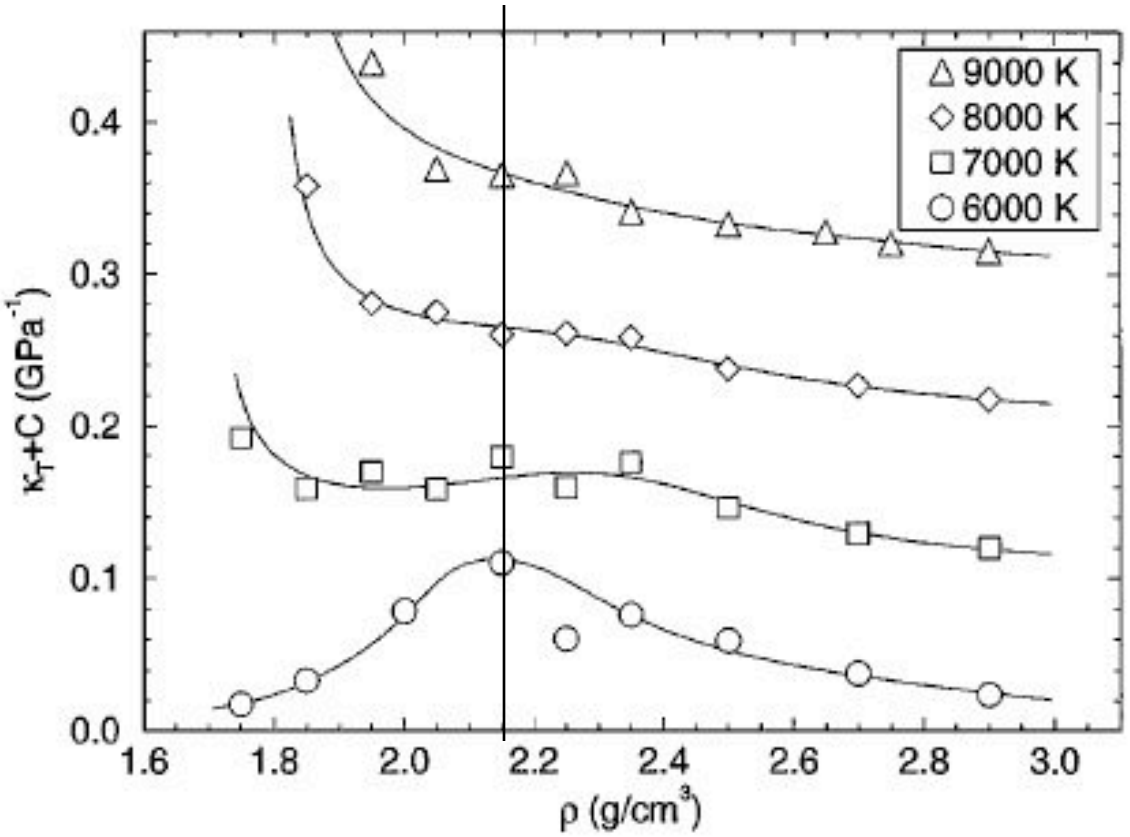


FIG. 4. Isotherms of κ_T as a function of ρ calculated using the derivative of the polynomial fits in Fig. 3 (solid lines), and using the piecewise slope of the P-ρ data in Fig. 3 (symbols). C is a shift parameter to facilitate comparison of the curves; $C = (T - 6000\ \mathrm{K})/(10^4\ \mathrm{K\,GPa})$.

Snapper rocks, Coolangatta, Qld.

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(fragile vs strong behavior)

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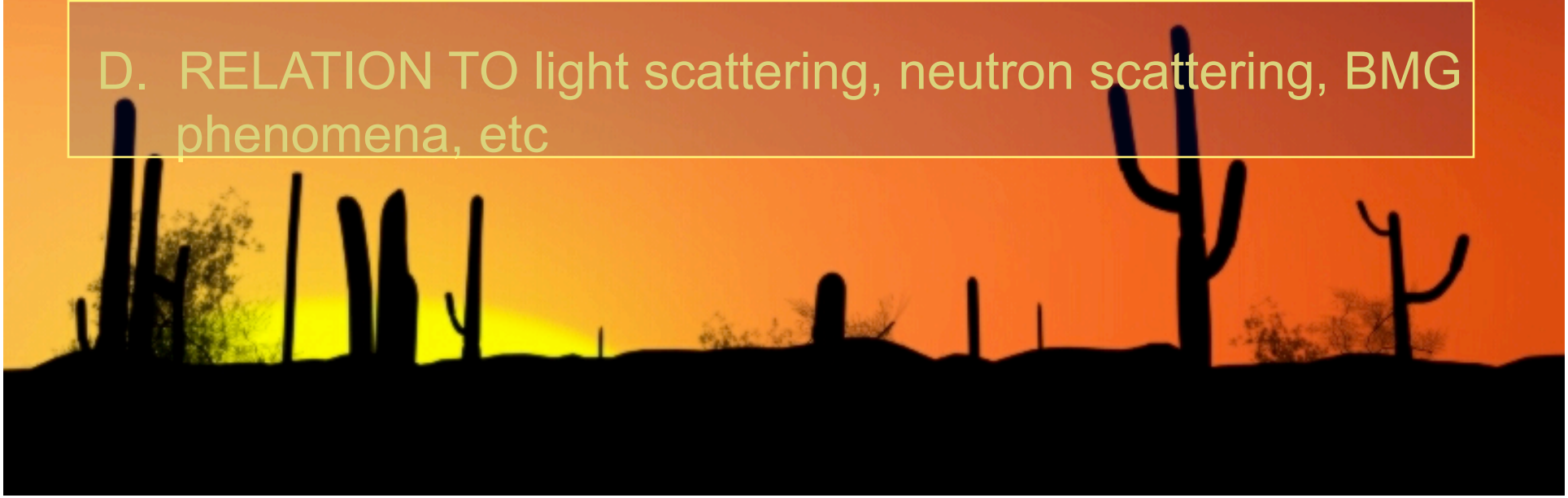


If we are right…

Then an important practical consequence is that the light scattering from silica fiberes should decrease when the fibre is annealed at lower temperatures.

Rayleigh scattering in relation to fictive temper-ature

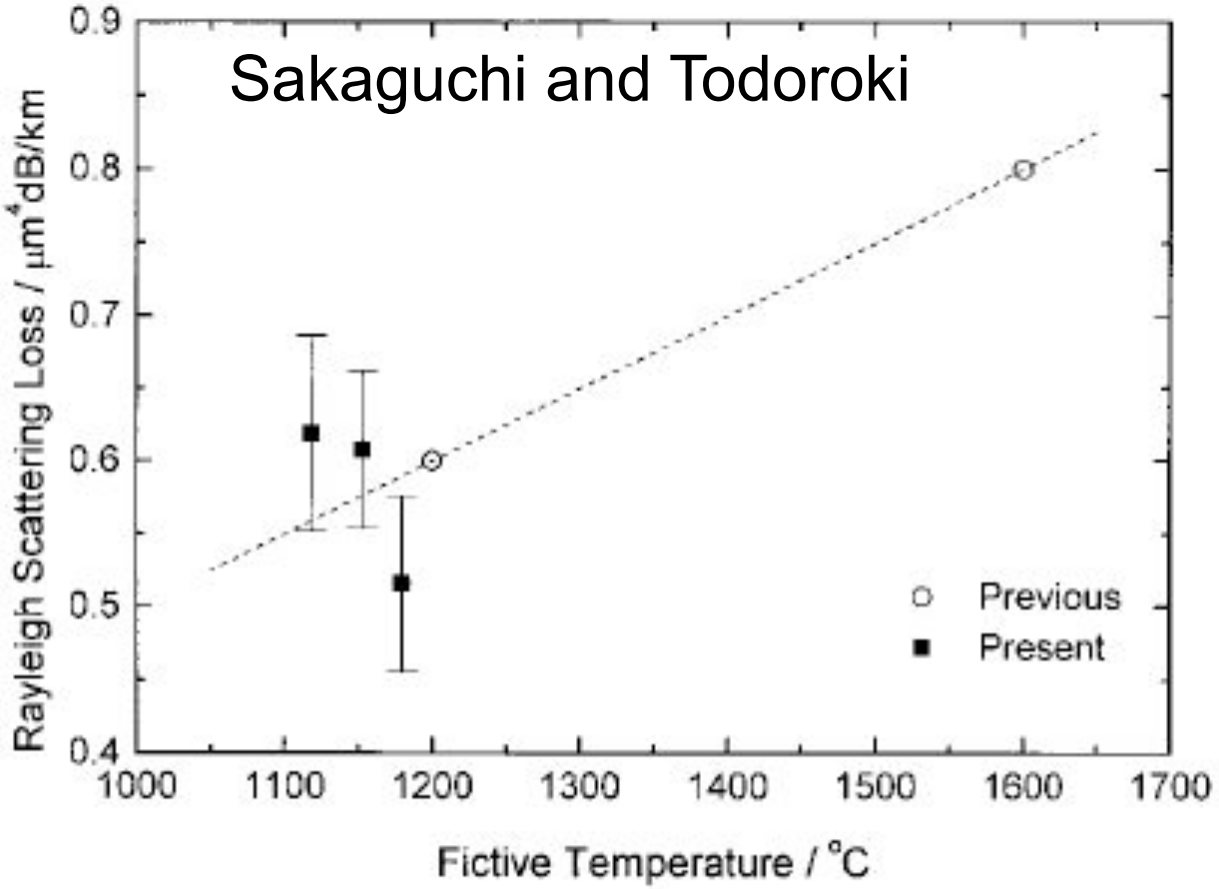


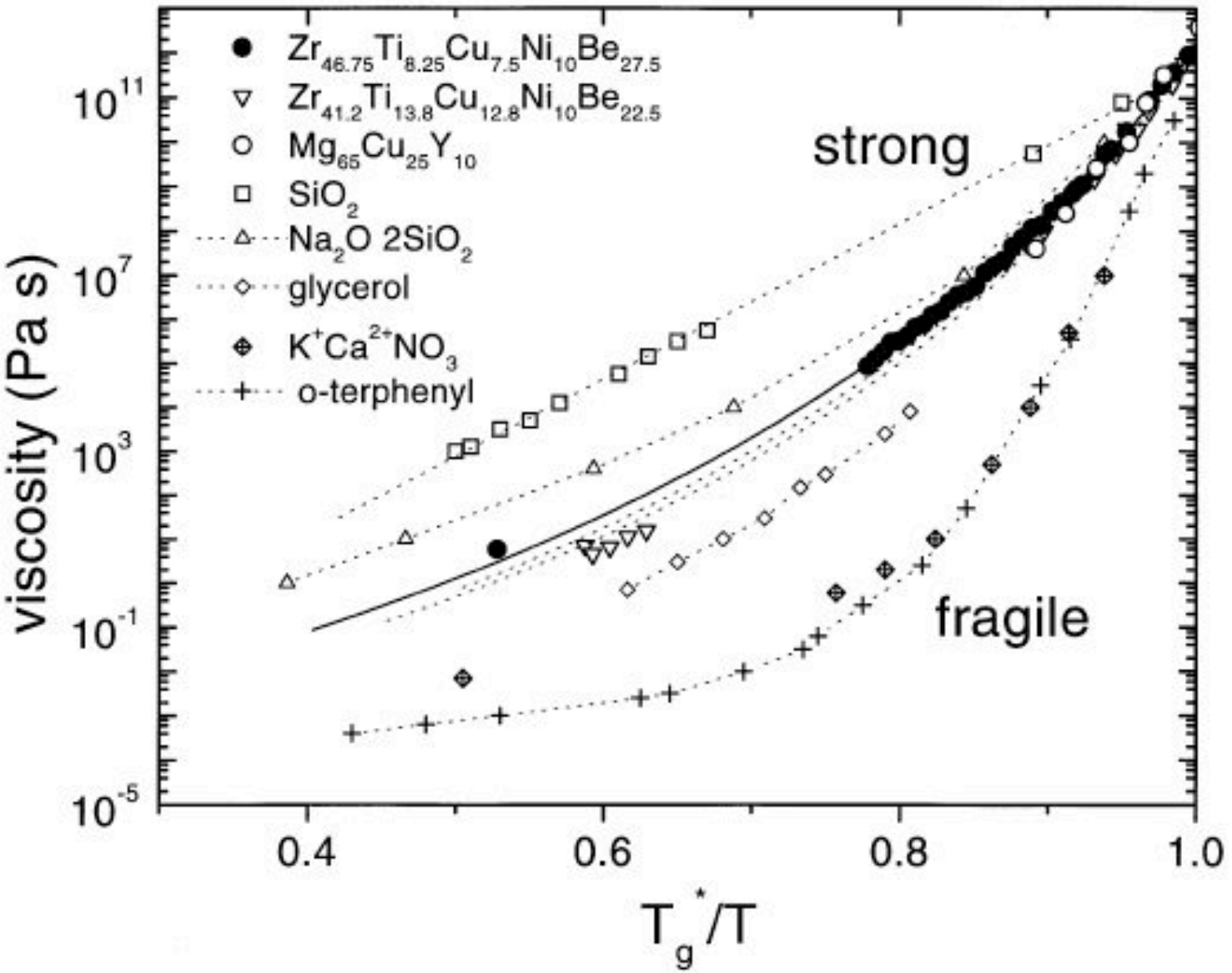
Fig. 5. Plot of the equilibrium scattering loss obtained from the data shown in Fig. 2 and the T_f of heated parts of the fiber. The dashed line was drawn based on the previous estimation, i.e., 0.8 μm^4 dB/km for silica core fiber with a T_f of 1600 °C and 0.6 μm^4 dB/km for silica glass with a T_f of 1200 °C.

"previous"

S. Sakaguchi and S. Todoroki, "Rayleigh scattering of silica glass and silica fibers with heat treatment," Jpn. J. Appl. Phys. Suppl. **37-1**, 56–58 (1998).

How about the BMG cases?

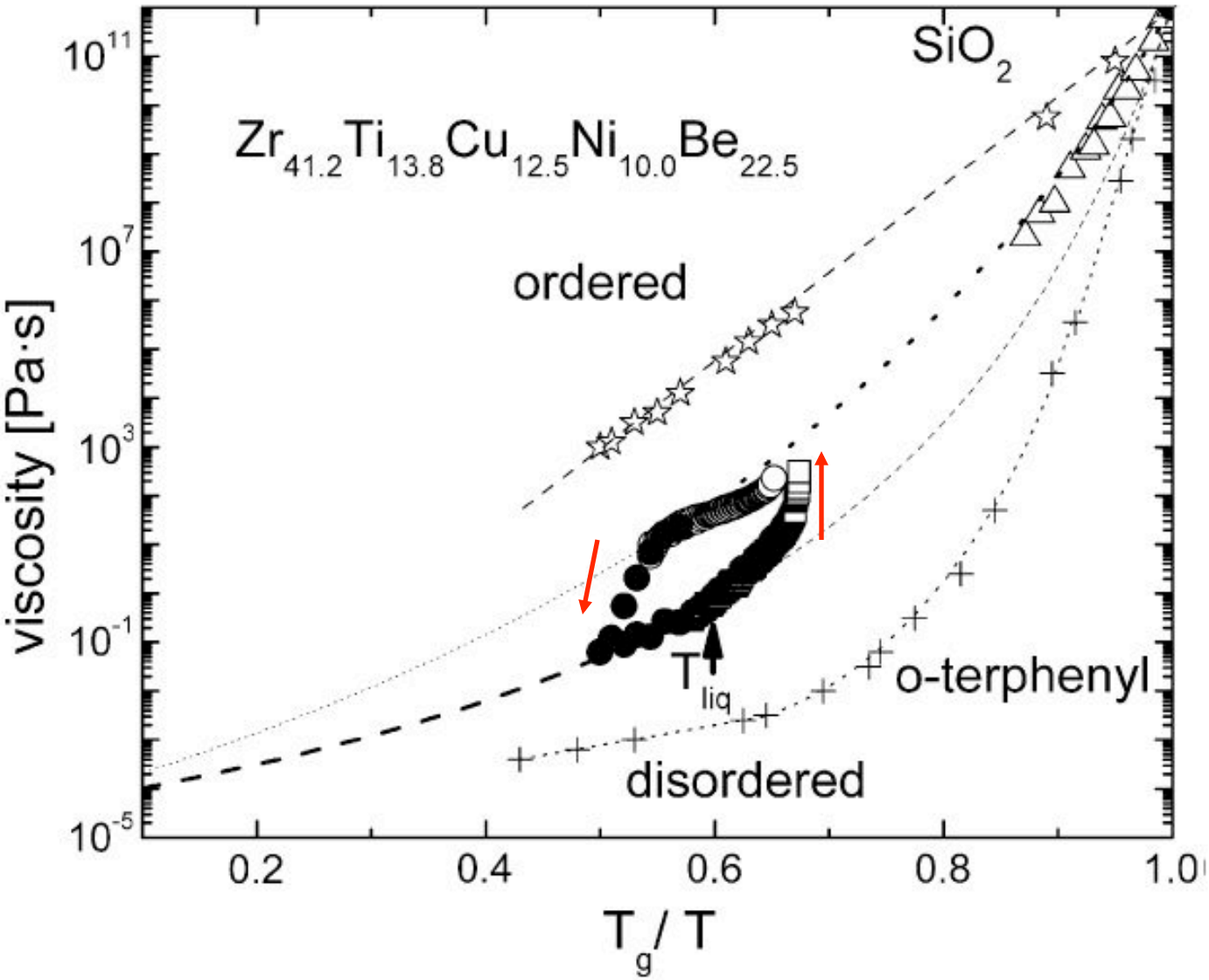
Busch Bakke and Johnson



But on holding the liquid at high temperature T >> Tg

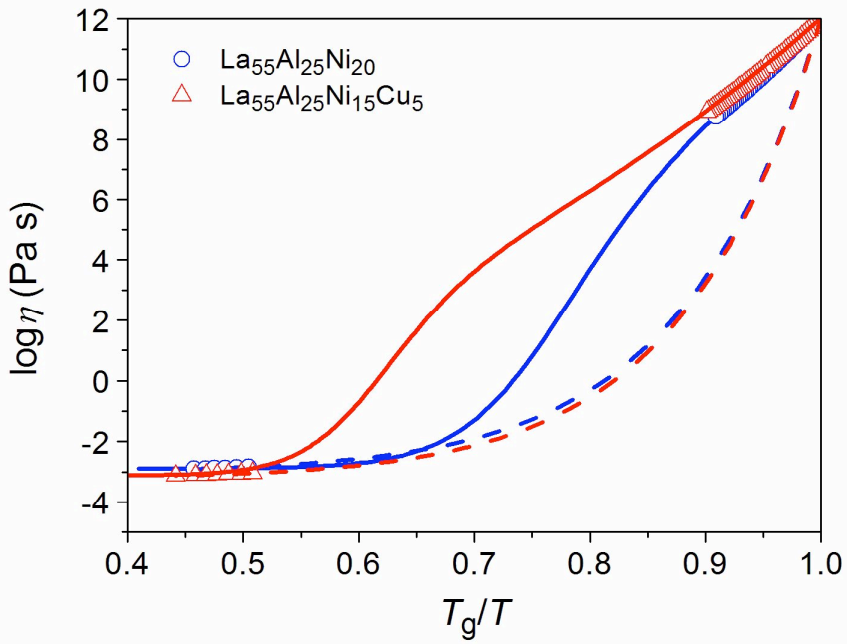
Ralf Busch and co-workers, Univ. Saarlandes

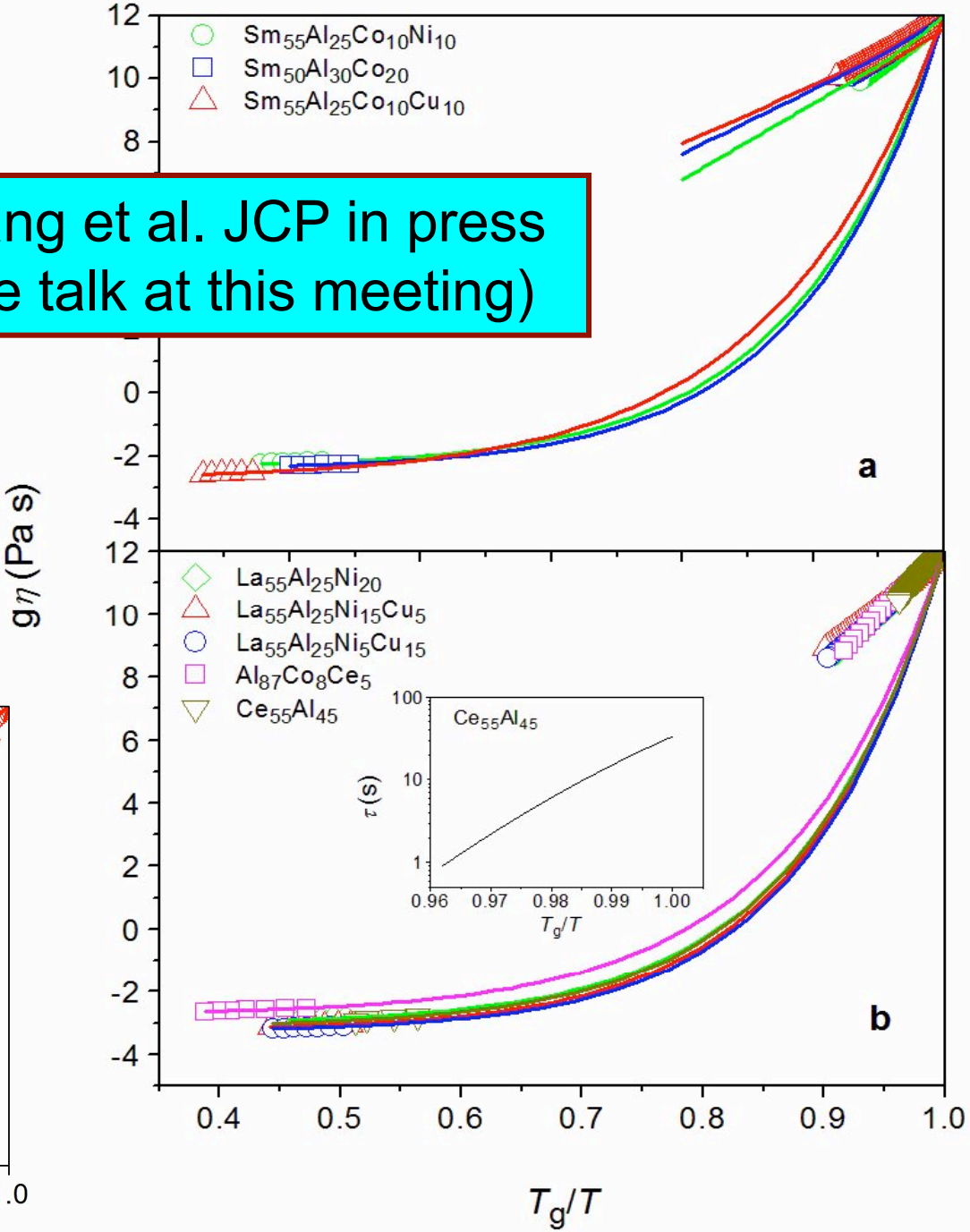
Looks like reversible depolymerization, after the fashion of liquid sulfur at around 160ºC. Entropy-driven break-up of linked polyhedra



Fragile-to-strong transition in metallic glassformers

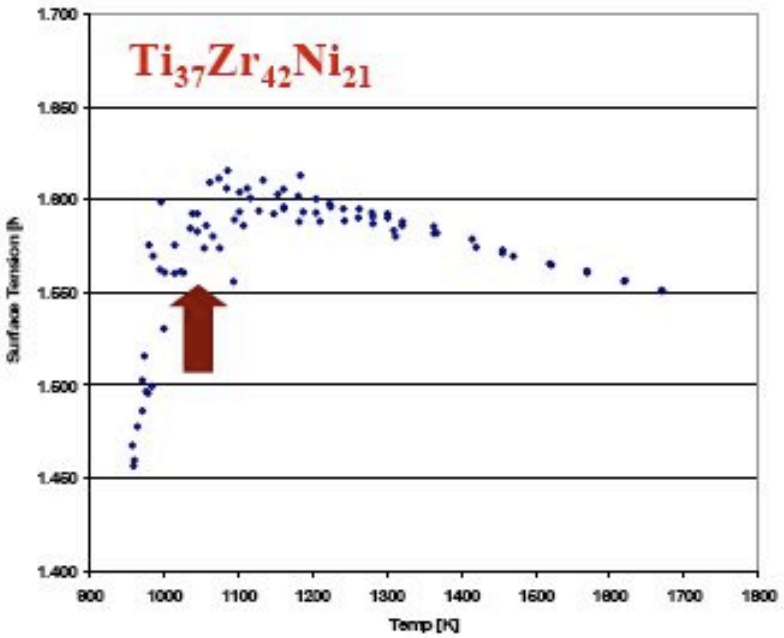
Zhang et al. JCP in press
(Yue talk at this meeting)





L-L transitions above Tg in BMG systems

Thermo- physical Properties of Supercooled Liquid

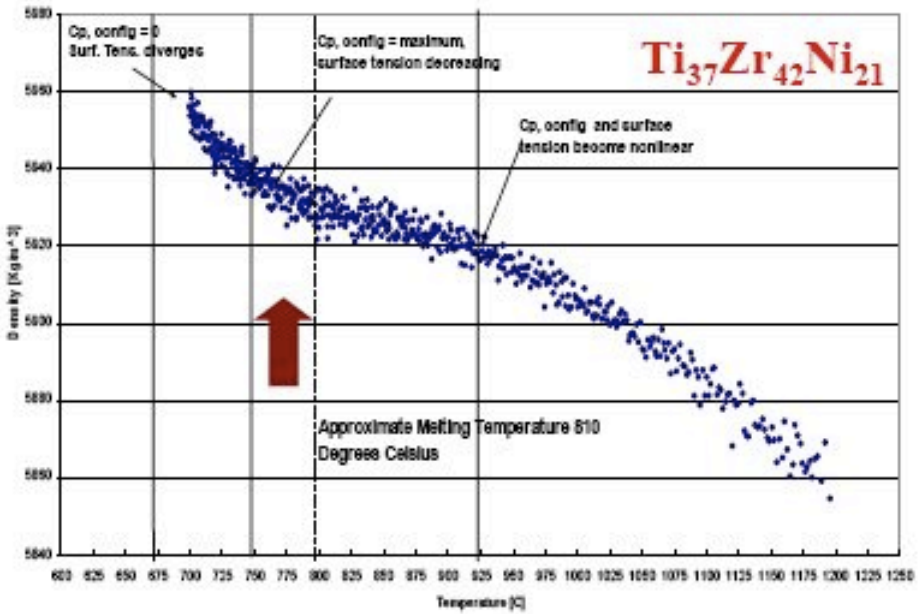


Thanks to Ken Kelton

(Washington U, St. Louis)

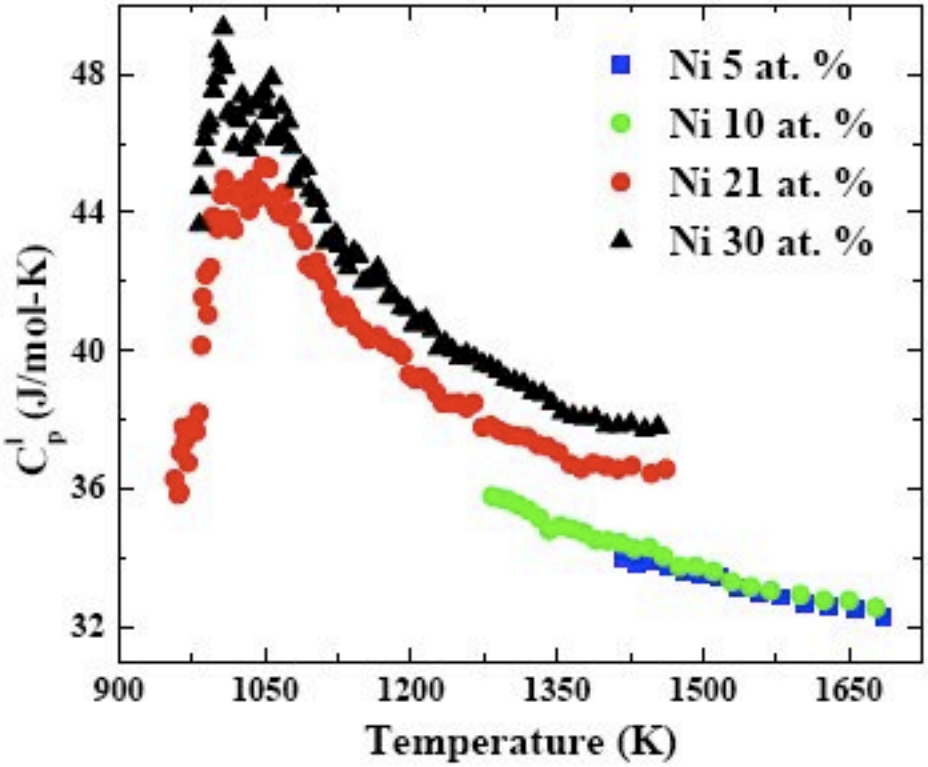
Dramatic decrease in surface tension

Density Inflection



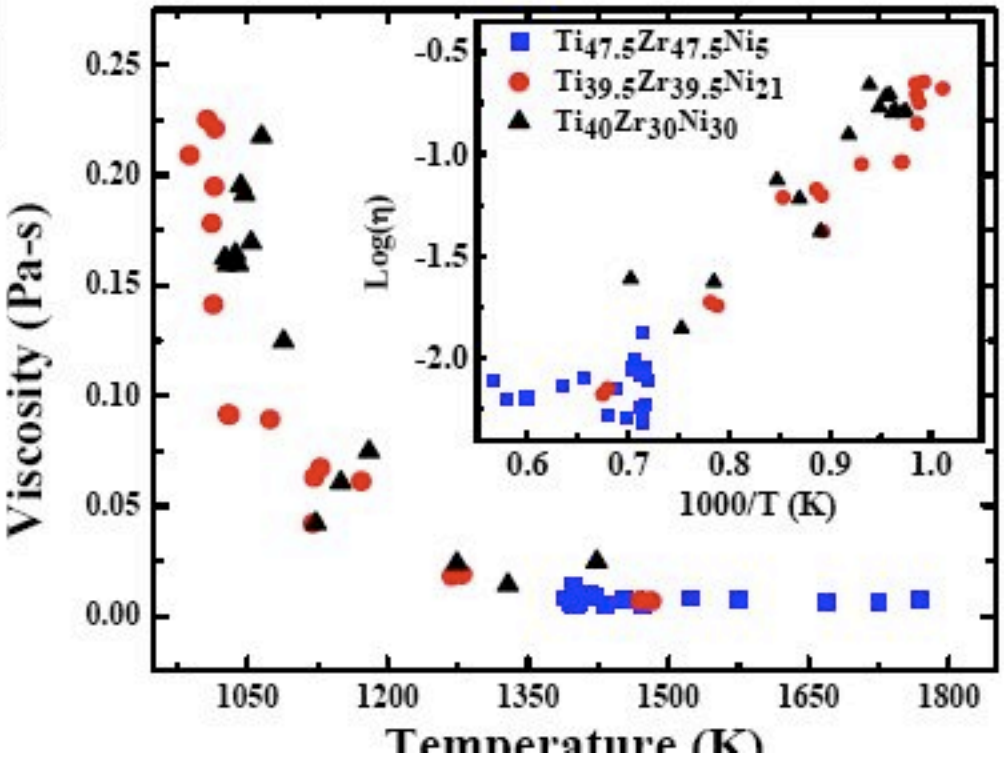
R. W. Hyers, R. Bradshaw, G. W. Lee,
A. Gangopadhyay, J. Rogers, K. F. Kelton.

Thermo- physical Properties of Supercooled Liquid



Possible specific heat maximum
Issues with noise, emissivity -
these are under investigation …

Ken Kelton (cont'd)

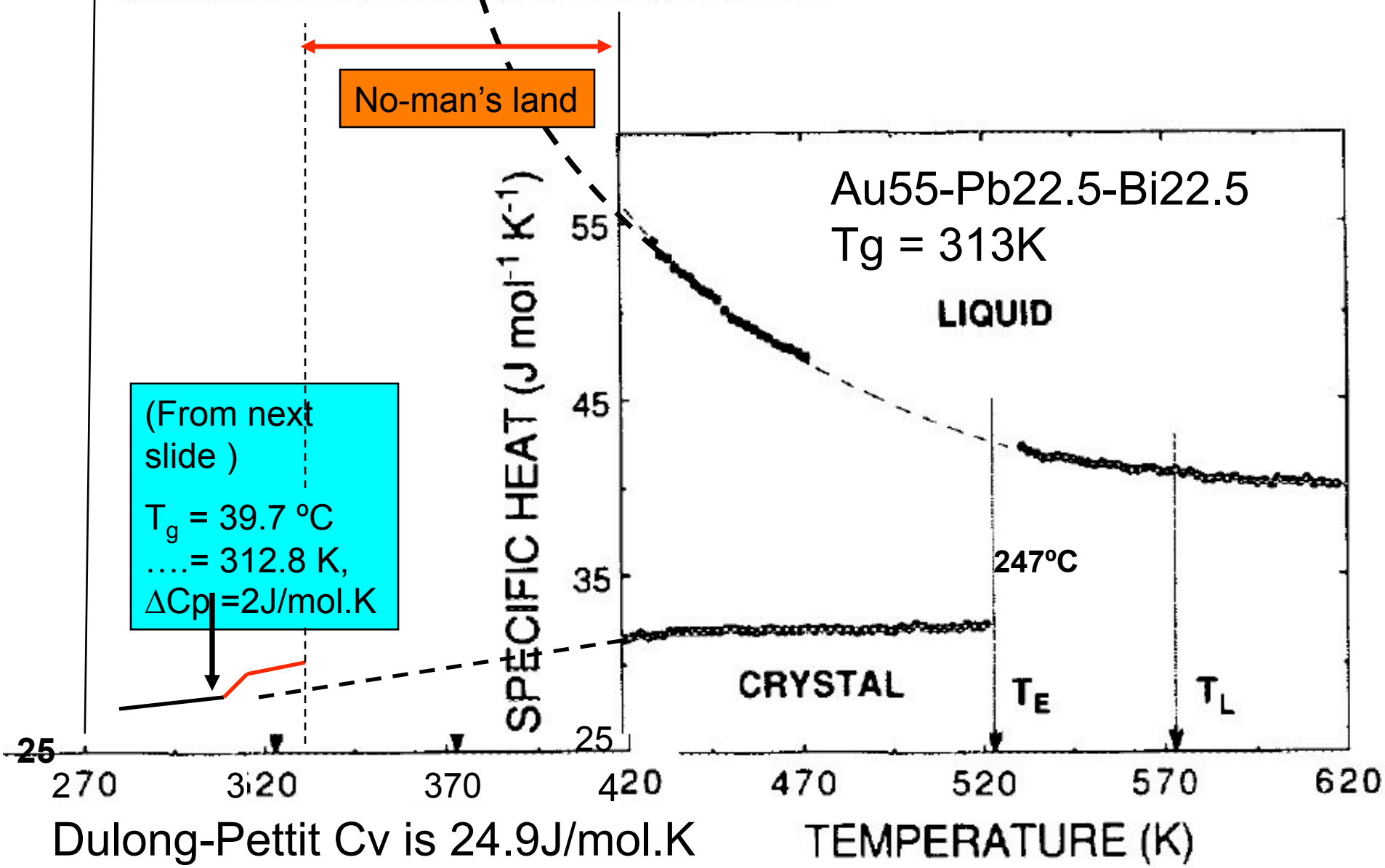


Rise in viscosity

R. W. Hyers, R. Bradshaw, G. W. Lee,
A. Gangopadhyay, J. Rogers, K. F. Kelton,

For T_g see:

M. C. Lee, H. J. Fecht, J. L. Allen, J. H. Perepezko, K. Ohsaka, and W. L. Johnson, J. Mater. Sci. Eng. 97, 301 (1988).



Dulong-Pettit Cv is 24.9J/mol.K

DSC thermogram for Au-Pb-Sb glass

Overshoot probably reflects ambient temperature (25ºC) annealing before the scan. The heat integral to 54ºC (0.78 J/g) could be turned into an average heat capacity increase above that of glass from 25ºC to 54ºC, of 0.79/29 J/g.K ≈ 0.03 J/g.K . This amounts to ~200 x 0.03 J/mole.K , i.e. ~2J/mol.-K, which is pretty tiny considered against 24.9 J/mol-K for the classical atomic solid. At T(eutec) (next slide), the excess heat capacity is already more than 10 J/ mol.K, so between the crystallization temperature and the glass temperature a huge Cp drop has apparently occurred. Why hasn't anyone brought this up for discussion, I wonder?

From Lee, Fecht, Perepezko, Johnson

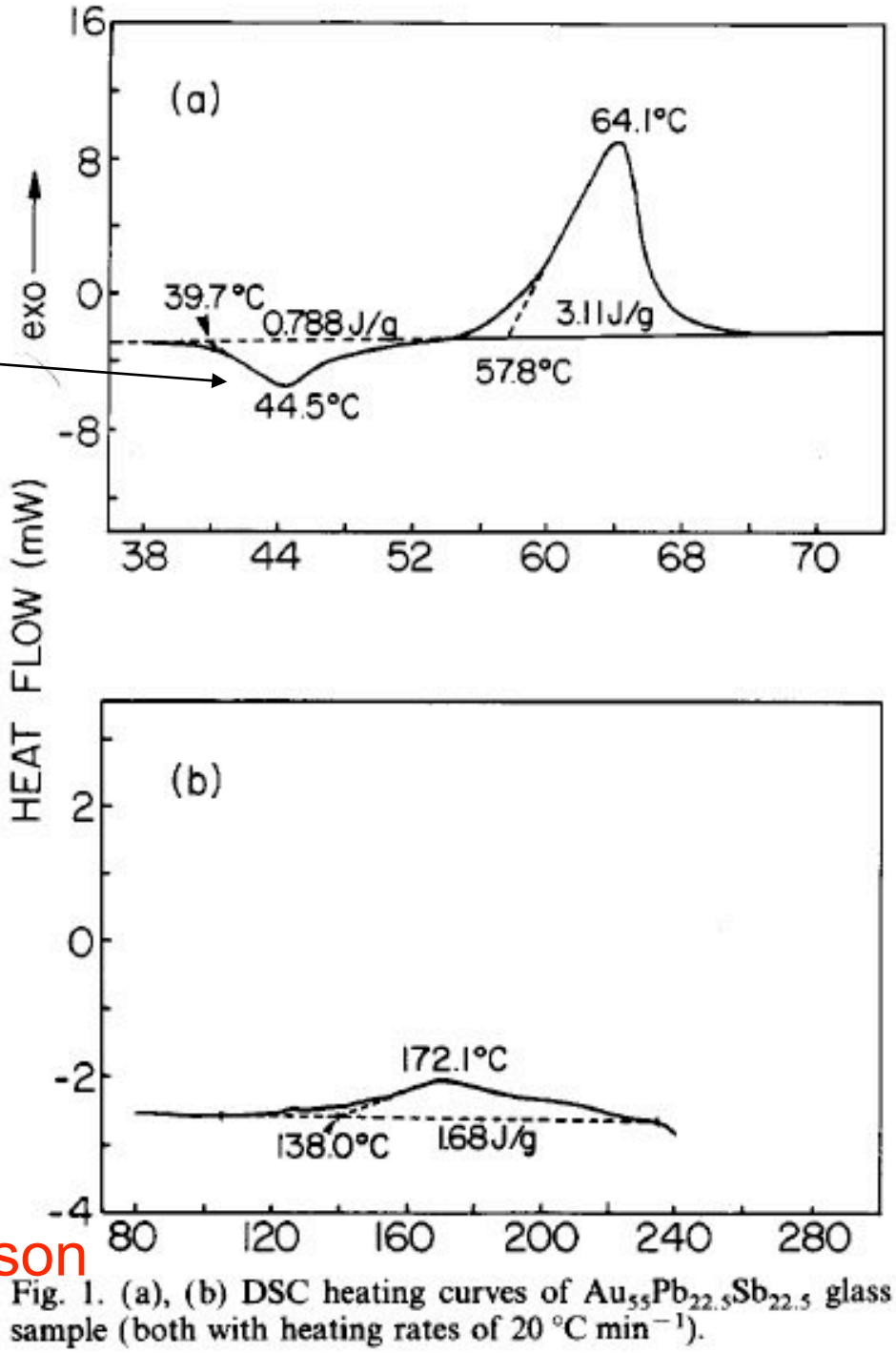
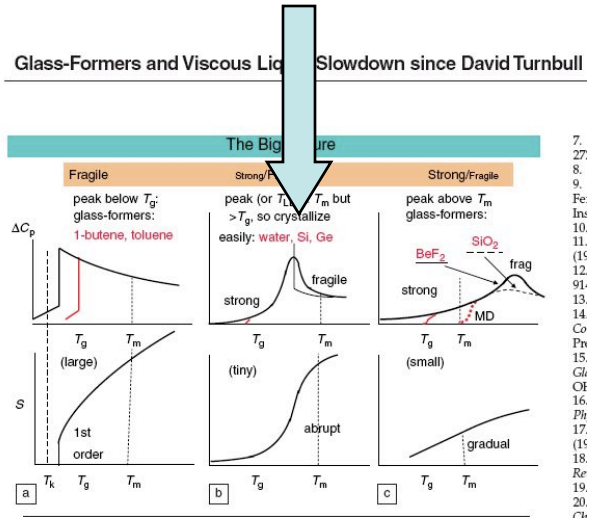


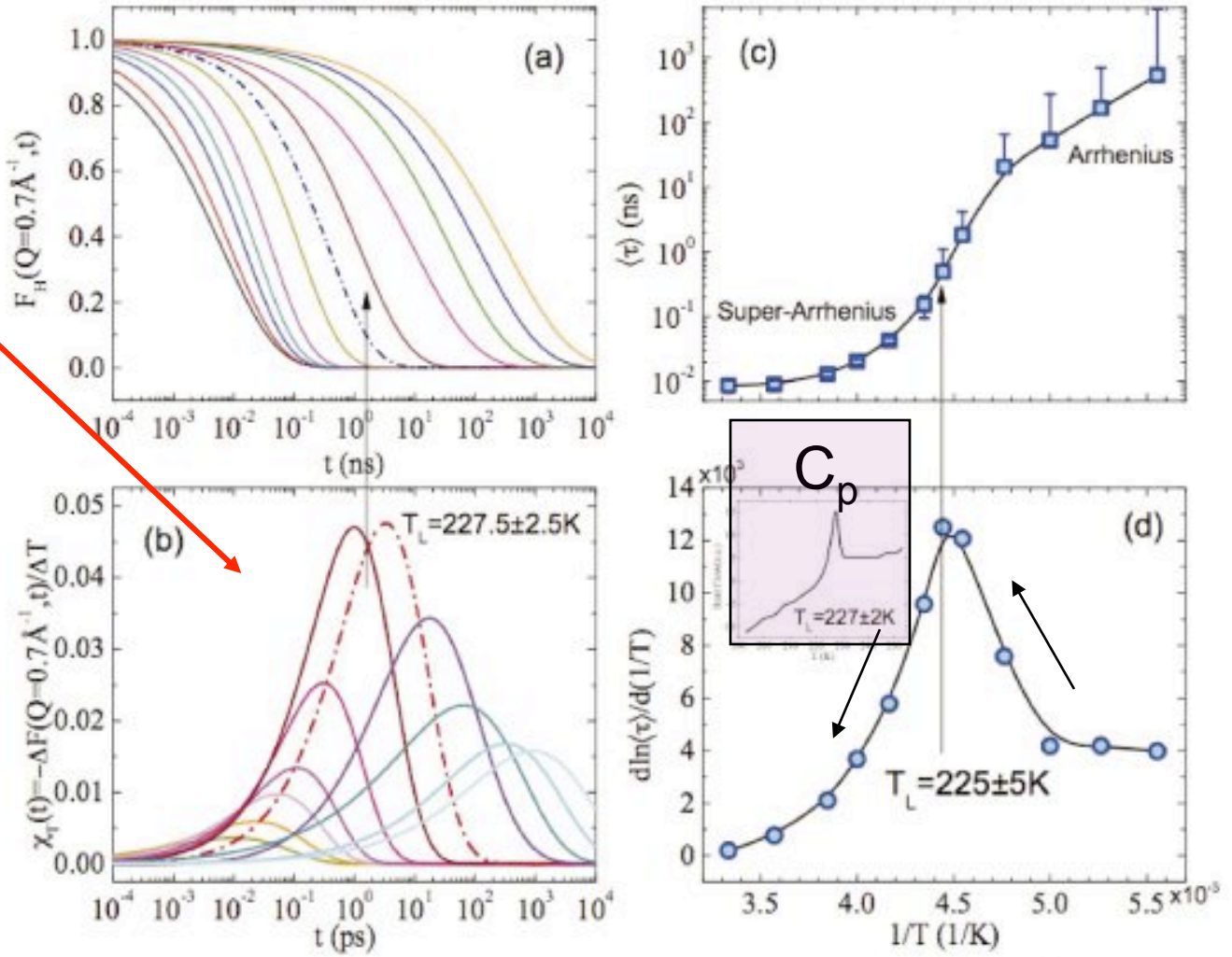
Fig. 1. (a), (b) DSC heating curves of $Au_{55}Pb_{22.5}Sb_{22.5}$ glass sample (both with heating rates of 20 °C min^{-1}).

How about (confined) water correlation length?

case the temperature change ΔT. $\chi_T(Q,t)$ also shows a peak at around τ(Q,T) when plotted as a function of time at constant Q, and the height of which is proportional to some sort of dynamic correlation length. Experimentally, $\chi_T(Q,t)$

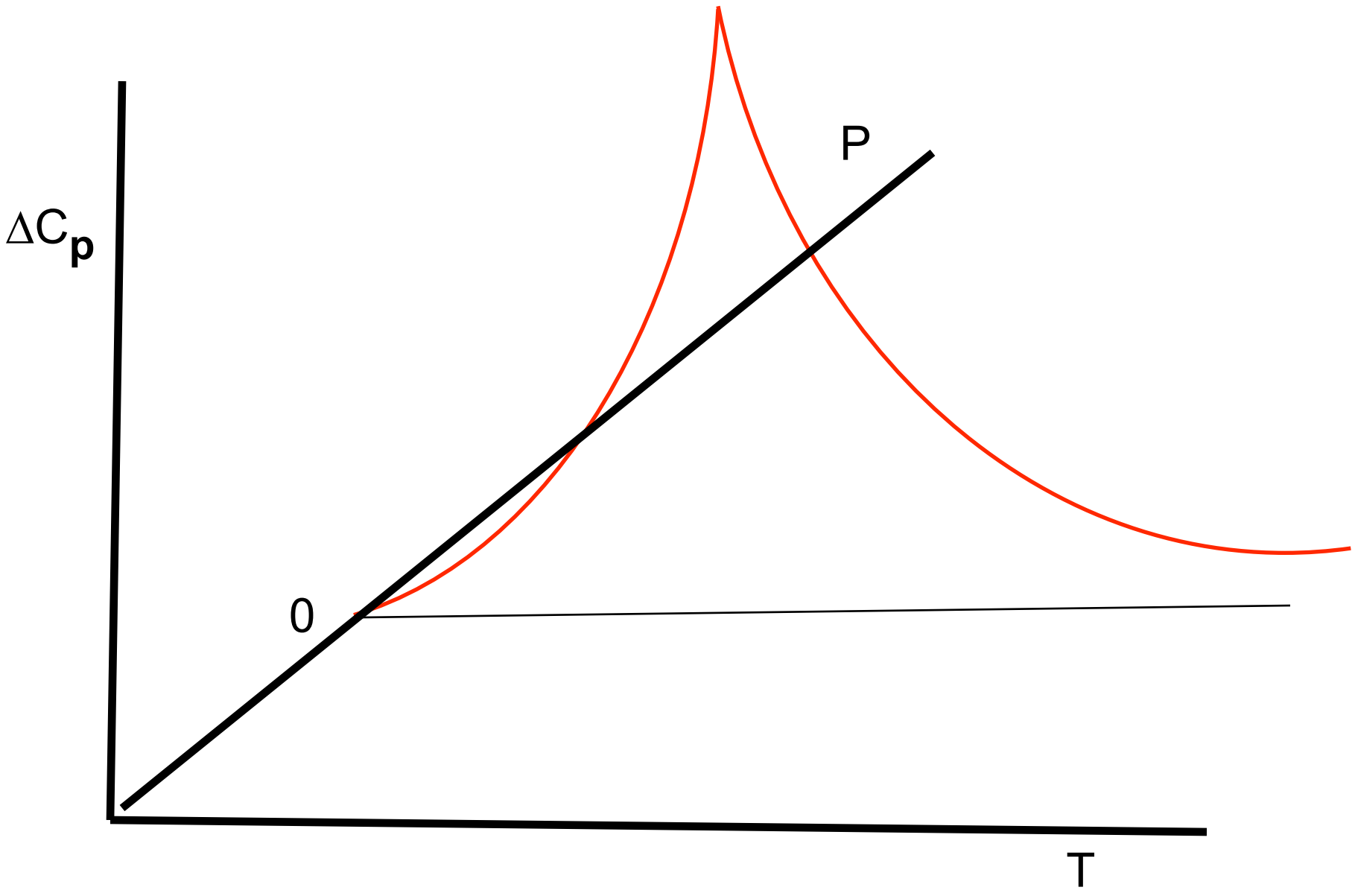
H.-S Chen et al. Dynamic response function, shows maximum correlation length at temperature of Cp peak





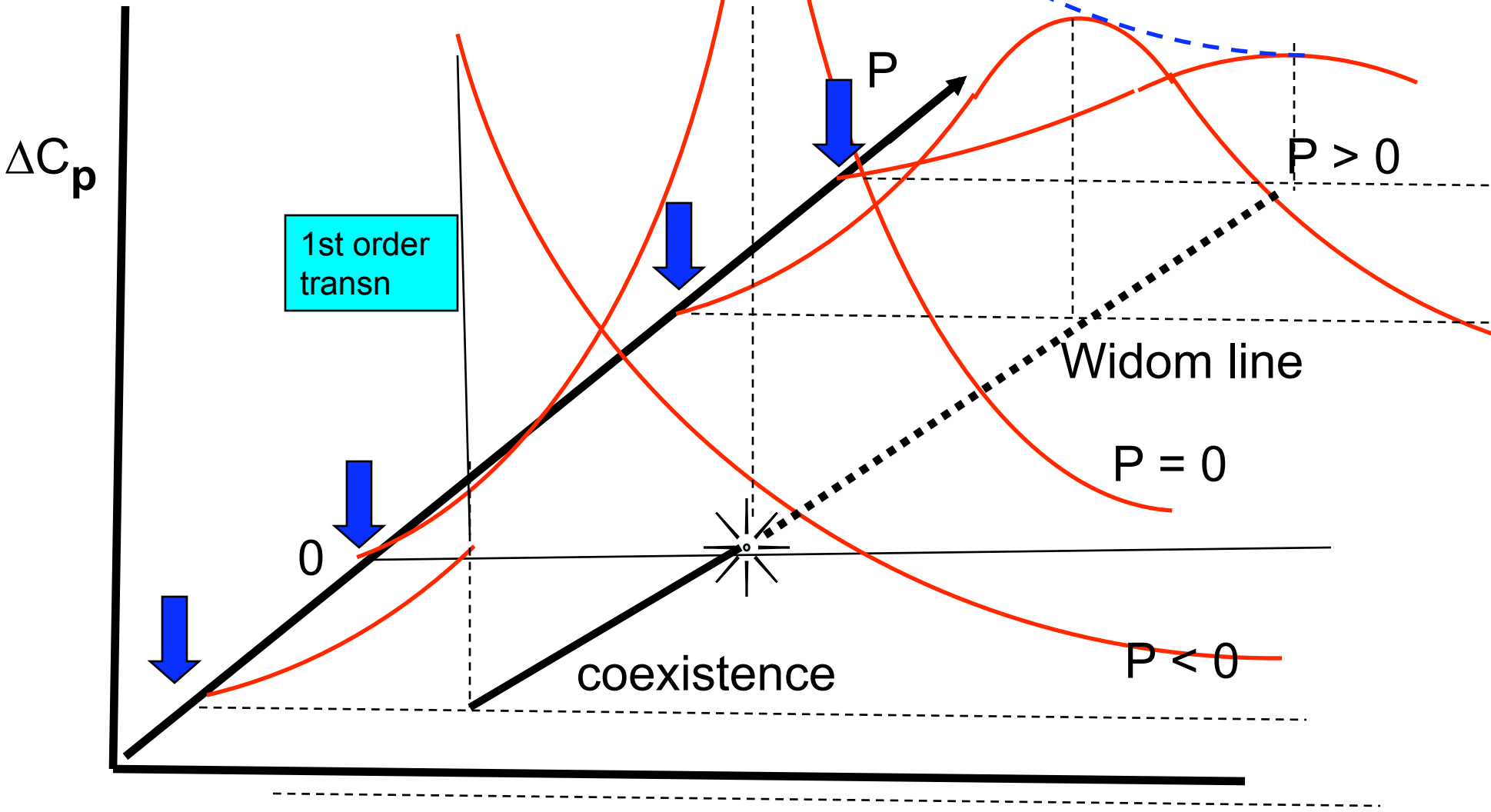
NOW let's look at how these different liquid state transitions can fit together in a comprehensive pattern. Look at what is expected "above" and "below" the lambda transition - that is a liquid-liquid critical point in the case of liquids

Lambda transition (Tc) at ambient



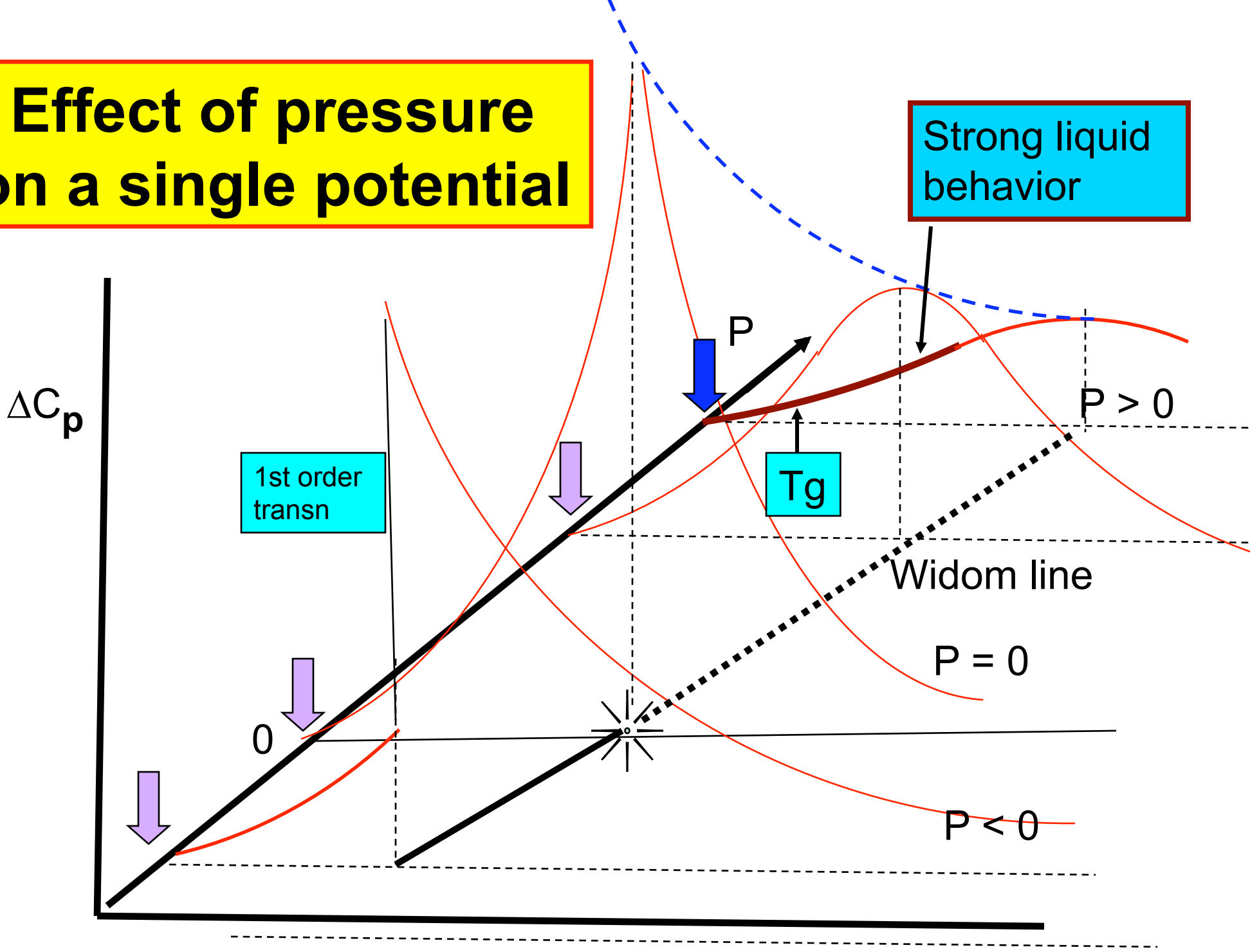
Effect of pressure on a single potential

Positive Clapeyron slope, p_c at p=0, (like metals, Jagla model)

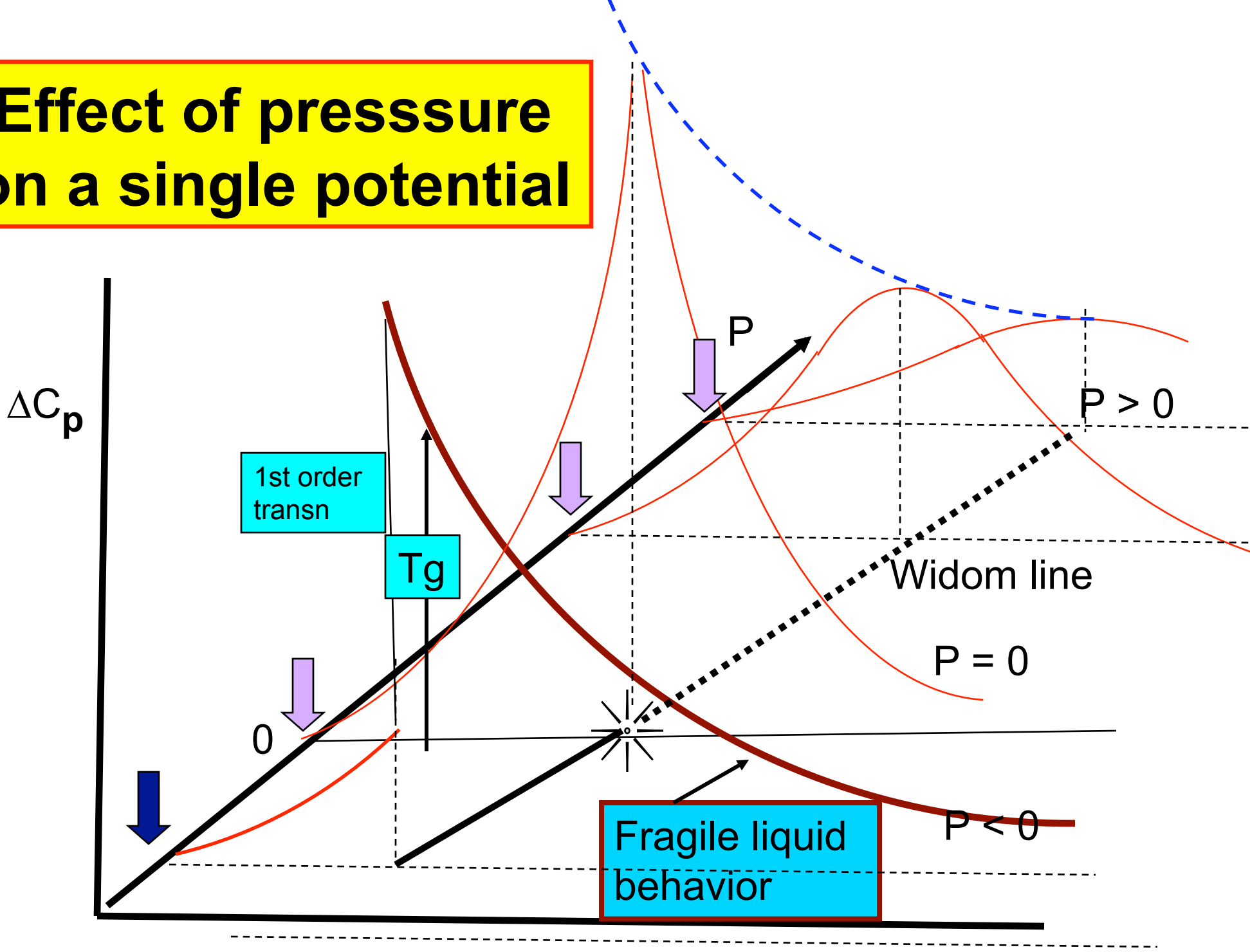


Let’s identify the strong and fragile elements again

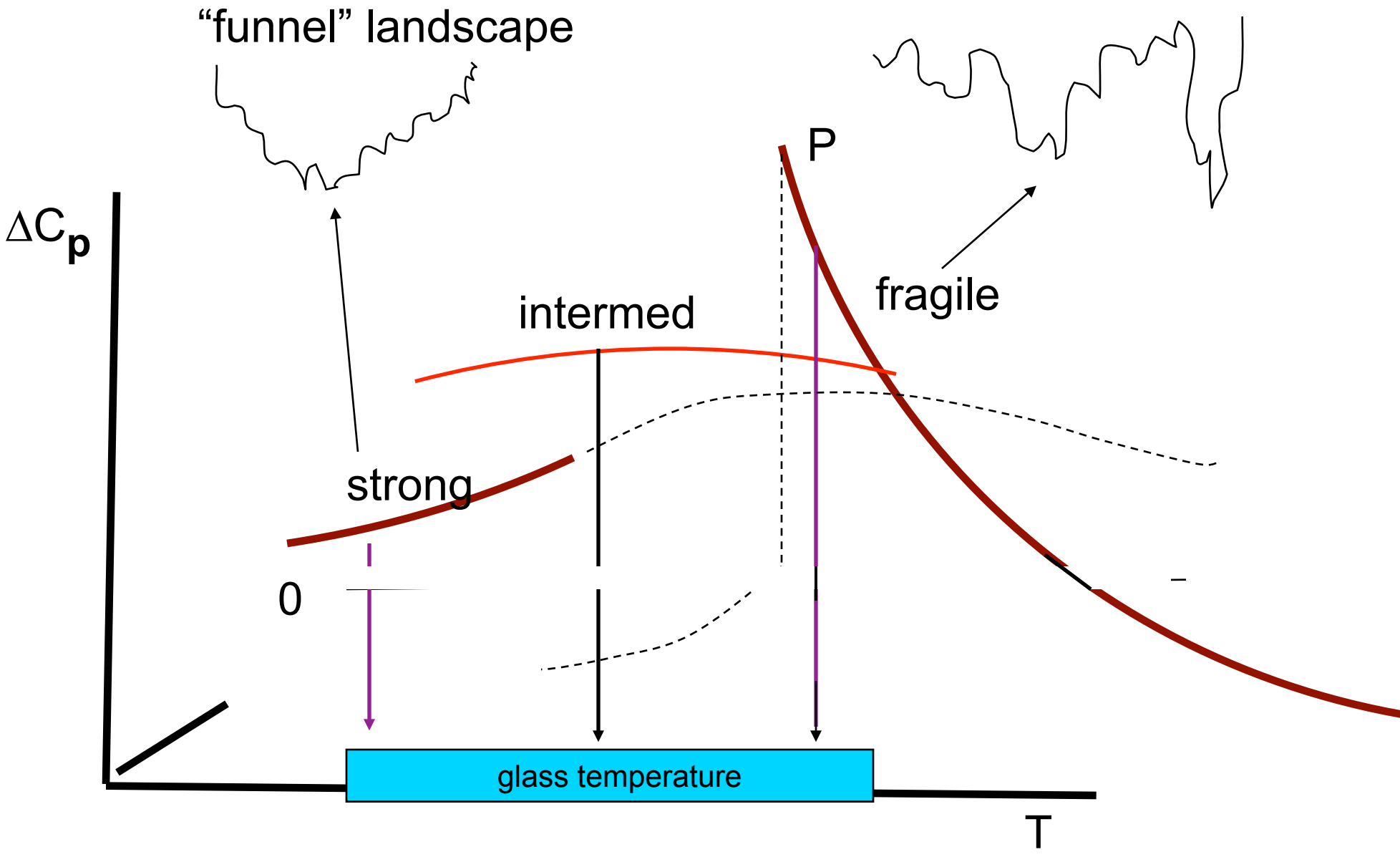
Effect of pressure on a single potential



Effect of presssure on a single potential



Strong liquid system accesses nearly all its landscape



Summary - liquids in equilibrium (>above T_g)

1. Understanding of fragility of GF is a major challenge.
2. It is as much a feature of the thermo, as of the kinetics.
3. Turn attention to correlation lengths, using insights from non-liquid "glassformers"
4. study S & F rotator phases, some end in lambda transitions, others melt.
6. study of metal alloy order-disorder transition: identify increasing correlation length going away from Tg.
7. Identify similarity to strong liquid behavior, and water at low temperature.
8. Looking at big picture. Water as Rosetta stone. Reversal of correlation length behavior across Widom line.
9. The fragile liquid end. Fitting excess heat capacity, and prediction of 1st order transition below Tg
10. Ultrastable glasses from vapor deposition below Tg
11. Tests of predictions (a) EB vs ECH (b) SiO_2