Fragility and its (proposed) correlation to other properties. What can we learn from high pressure experiments?

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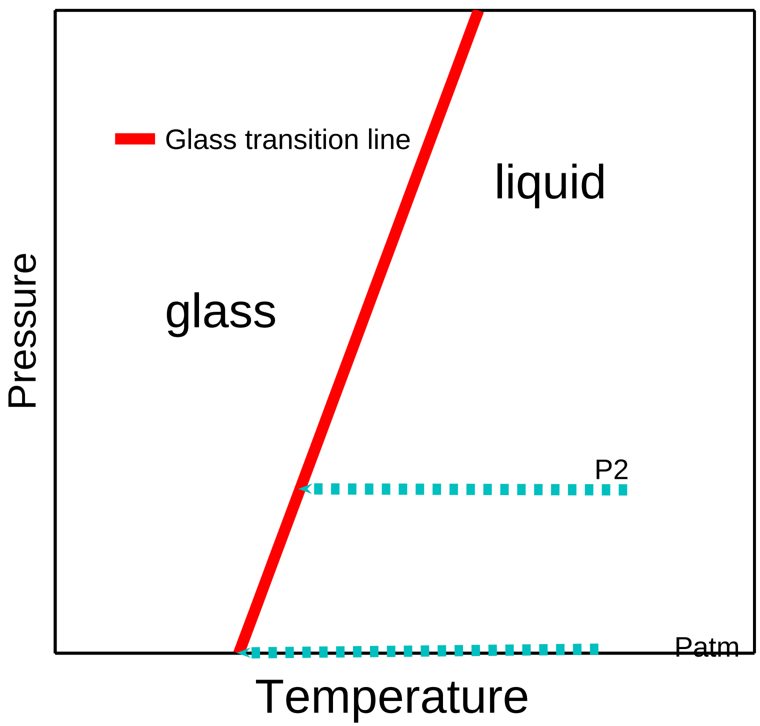
Outline

- Fragility and pressure
  - isochoric fragility
  - density scaling
- Correlations and pressure
  - $f_Q(T_g)$
  - β_{KWW}

Effect of pressure

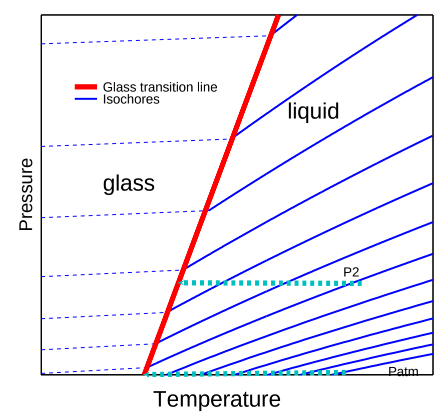
$$m_P = \left.\frac{\partial \log_{10}(\tau_\alpha)}{\partial T_g/T}\right|_P (T=T_g)$$

$$\tau_\alpha(T_g) = \tau_g$$

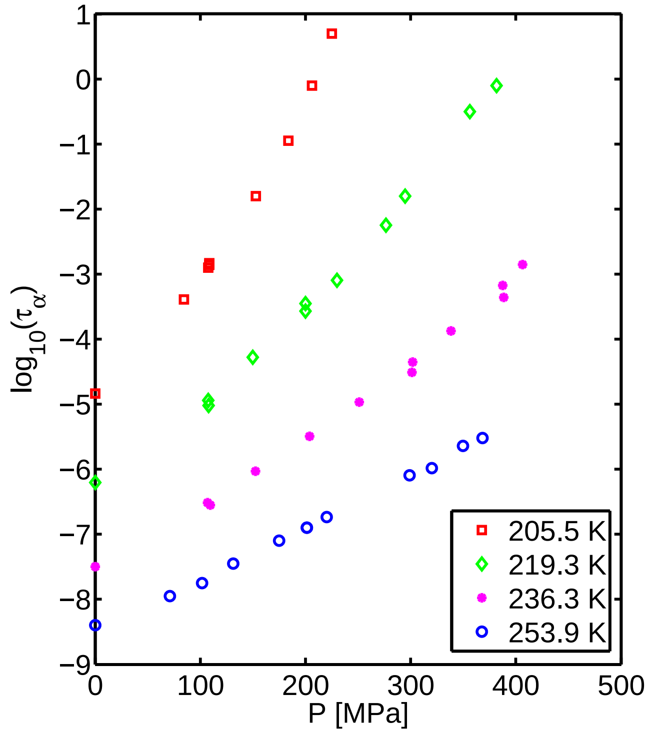


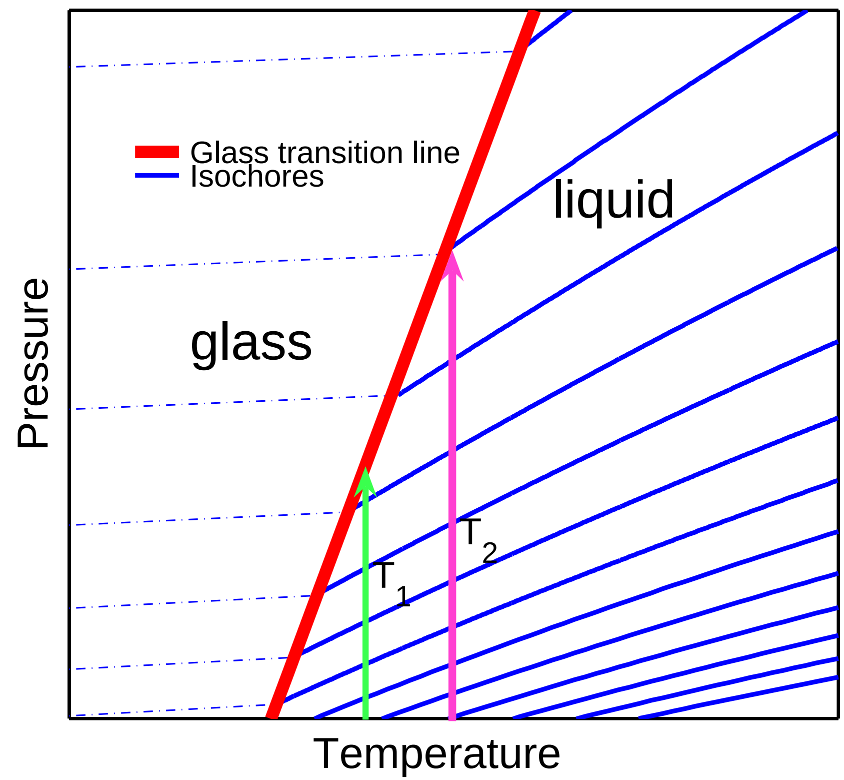
Isobaric glass transition
P>Patm

- higher T_g
- different (smaller) fragility
- same relaxing entity



Isothermal glass transition





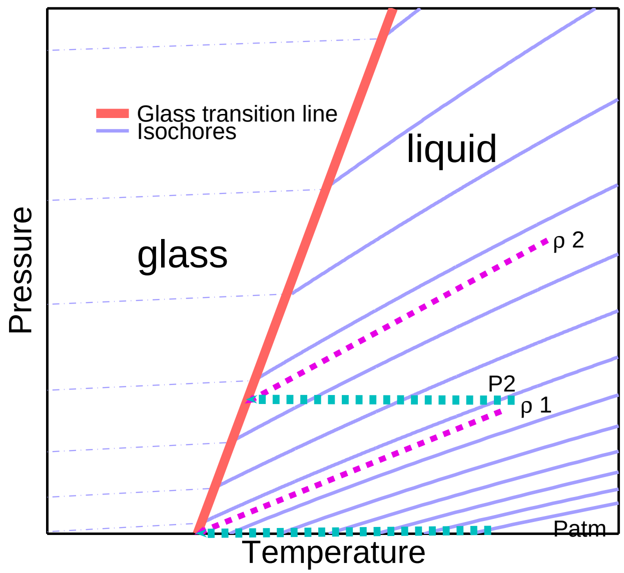
τ_α of DBP

4 different isotherms

dielectric spectroscopy
Niss *et al.* Phys. Cond. Mat. (2007)

Forming a glass at constant temperature.

Isochoric glass transition

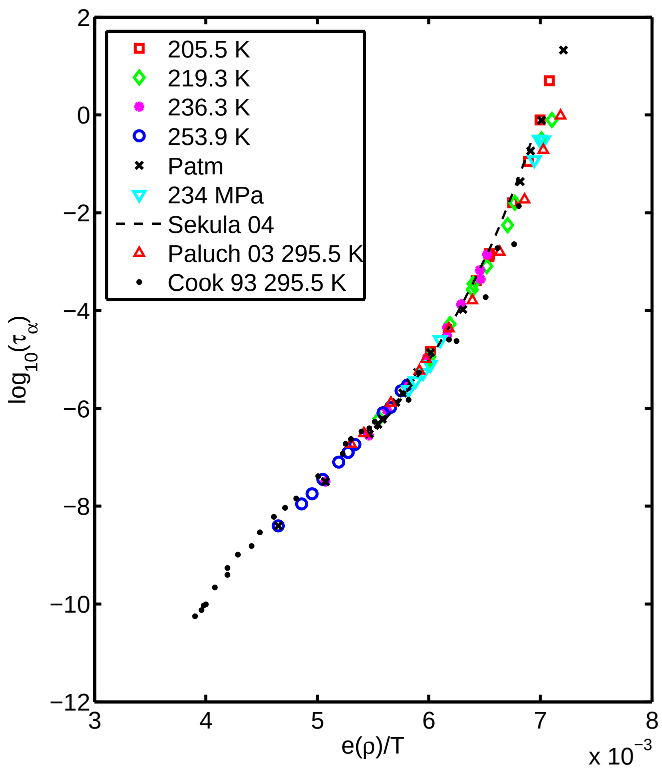


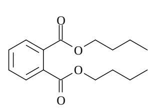
Forming a glass at constant volume

$$m_\rho = \left.\frac{\partial \log_{10}(\tau_\alpha)}{\partial T_g/T}\right|_\rho (T{=}T_g)$$

$$\begin{aligned} m_P &= \left.\frac{\partial \log_{10}(\tau_\alpha)}{\partial T_g/T}\right|_\rho + \left.\frac{\partial \log_{10}(\tau_\alpha)}{\partial \rho}\right|_T \left.\frac{\partial \rho}{\partial T_g/T}\right|_P \\ &= m_\rho + \left.\frac{\partial \log_{10}(\tau)}{\partial \rho}\right|_T \left.\frac{\partial \rho}{\partial T_g/T}\right|_P \\ &= m_\rho (1 + \alpha_P / |\alpha_\tau|) \end{aligned}$$

Scaling



DBP 

$$\tau_\alpha(\rho, T) = \tau_0 \exp\left(\frac{E(\rho,T)}{T}\right)$$

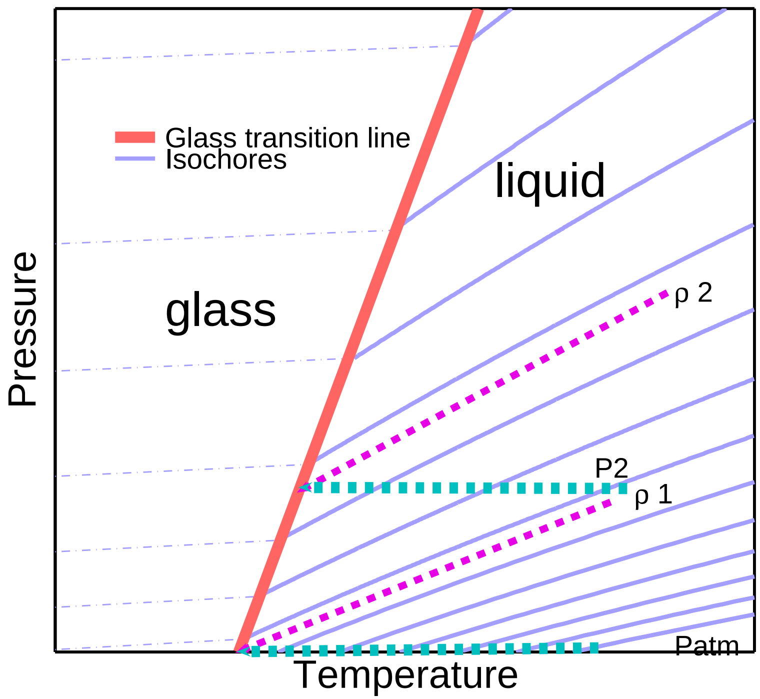
$$\frac{E(\rho,T)}{e(\rho)} = \Phi\left(\frac{T}{e(\rho)}\right)$$

$$\tau_\alpha(\rho, T) = F\left(\frac{e(\rho)}{T}\right), \quad e(\rho) = \rho^x$$

Alba-Simionesco, J. Chem. Phys. (2002)
Dreyfus, Eur. Phys. J. B (2004)
Roland, Rep. Prog. Phys. (2005)
Reiser, Phys. Rev. B (2005)

Niss, J. Phys. Cond. Mat. (2007)

Isochoric glass transition



Forming a glass at constant volume

If scaling holds then

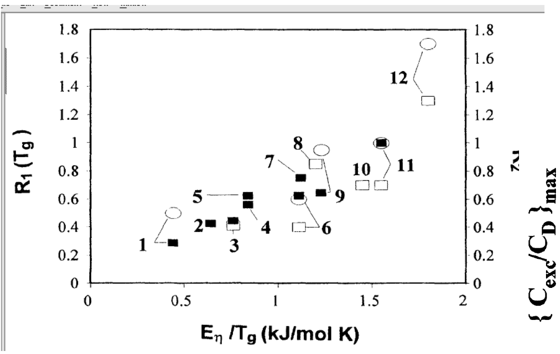
m_ρ : constant

$$m_P = m_\rho (1 + \alpha_P / |\alpha_\tau|)$$

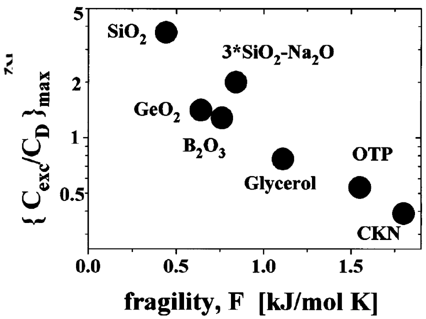
$$m_P = m_\rho \left(1 + \alpha_P T_g \frac{\mathrm{d} \log e(\rho)}{\mathrm{d} \log \rho} \right)$$

$$m_P = m_\rho (1 + \alpha_P T_g x)$$

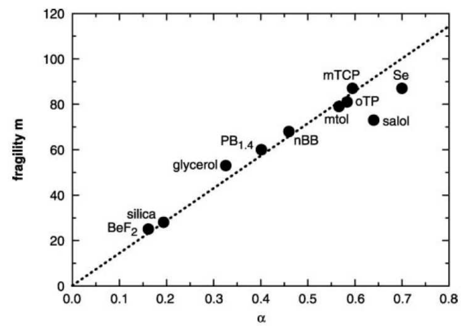
Correlations



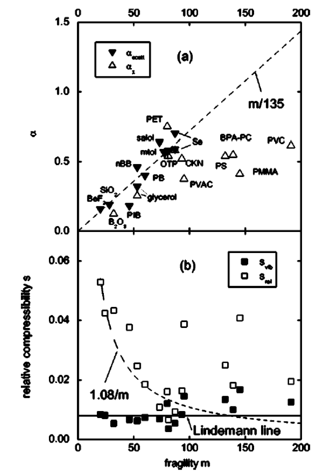
Sokolov et *al.* 1993



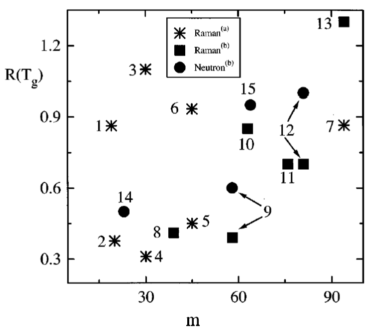
Sokolov et *al.* 1997



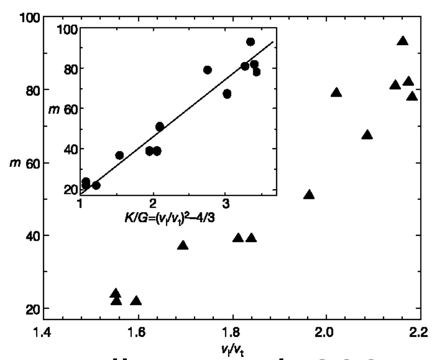
Scopigno et *al.* 2003



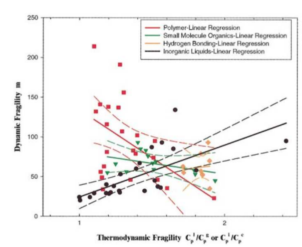
Buchenau et *al.* 2004



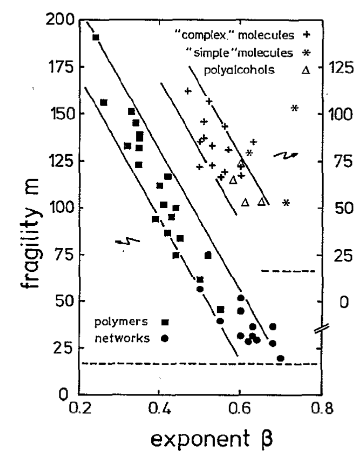
Yannopoulos et *al.* 2000



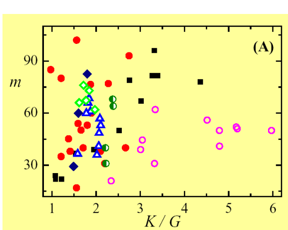
Novikov et *al.* 2004



Huang et *al.* 2000



Böhmer et *al.* 1993

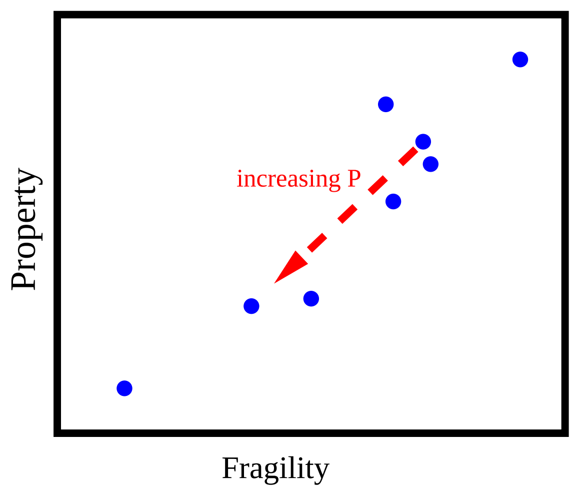


Yannopoulos et *al.* 2006

Pressure and correlations

isobaric fragility changes

are correlations robust ?



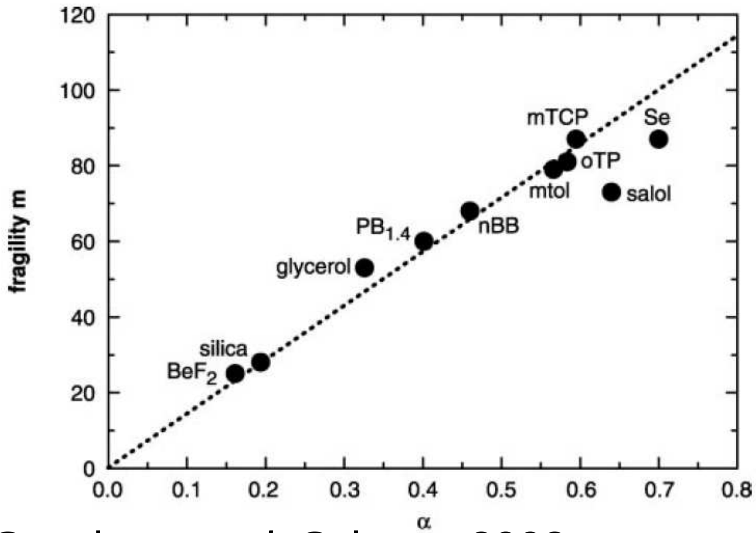
$$m_P(P) = m_\rho \left(1 + \alpha_p T_g \frac{\mathrm{d}\log e(\rho)}{\mathrm{d}\log \rho}\right)$$

m_ρ
constant
effect of temperature

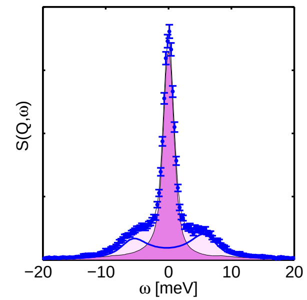
$\alpha_p T_g \frac{\mathrm{d}\log e(\rho)}{\mathrm{d}\log \rho}$
pressure dependent
effect of density

Niss and Alba-Simionesco, Phys. Rev B, (2006)

$f_q(T_g)$

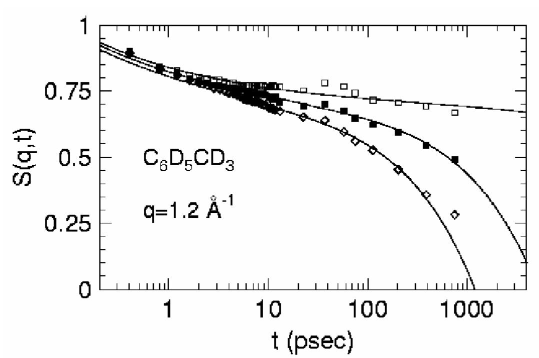


Scopigno et *al.* Science 2003

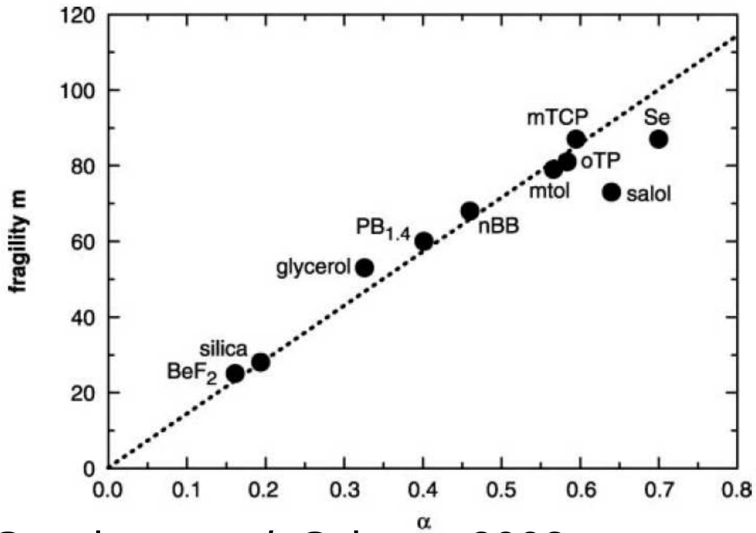


$Q = 2\text{nm}^{-1}$

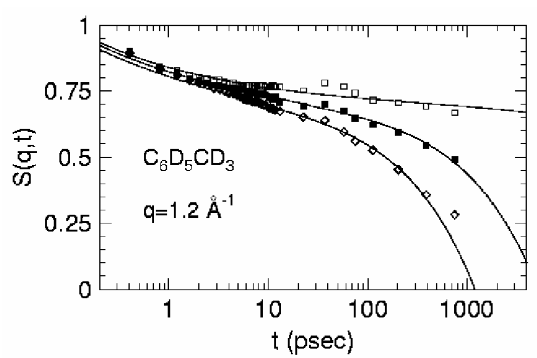
IXS ESRF

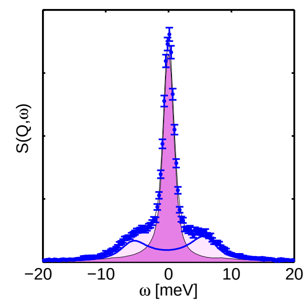


$f_q(T_g)$



Scopigno et *al.* Science 2003





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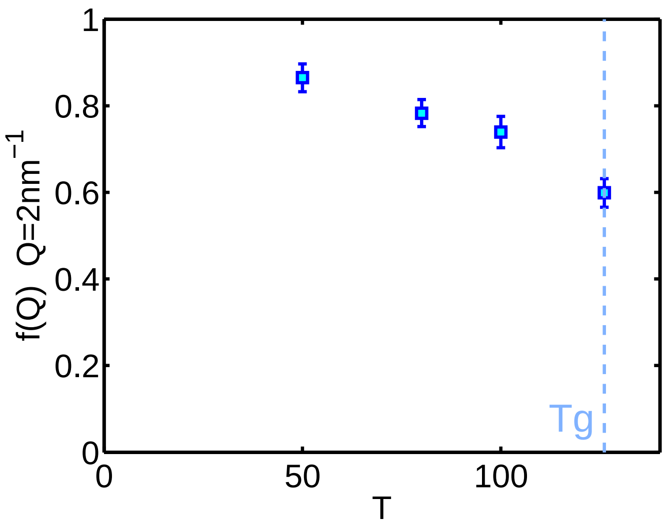
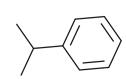
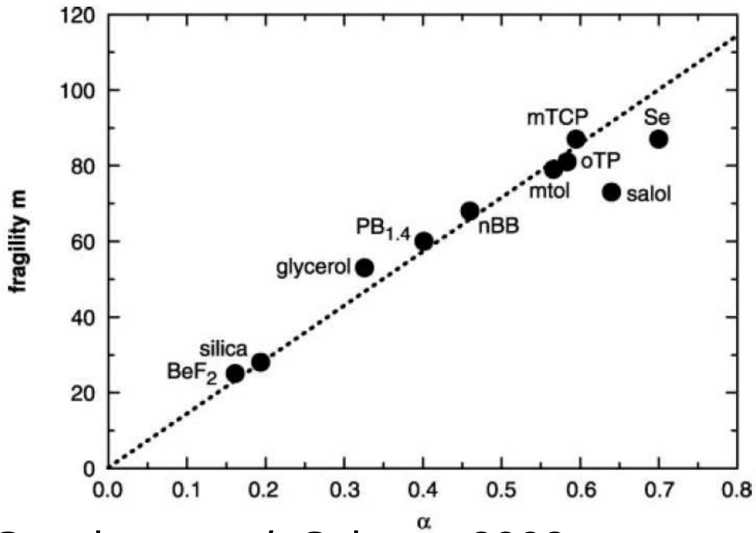
IXS ESRF

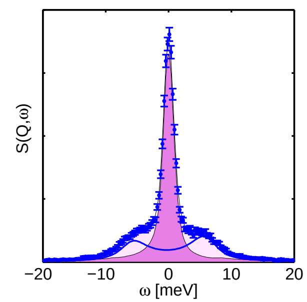


Cumene  ID16

$f_q(T_g)$

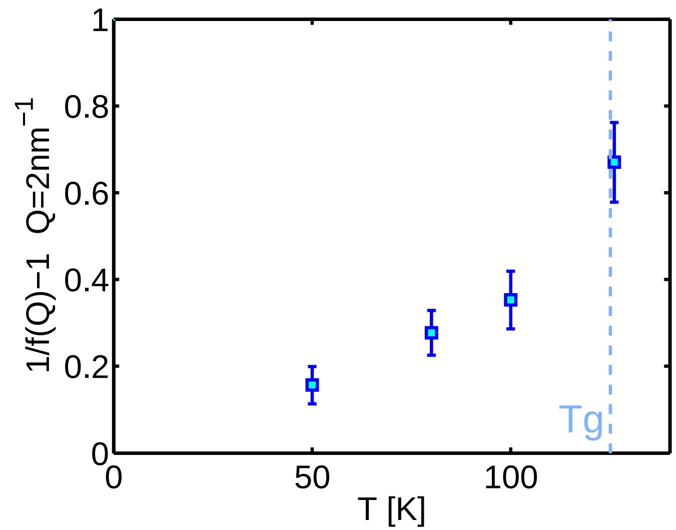


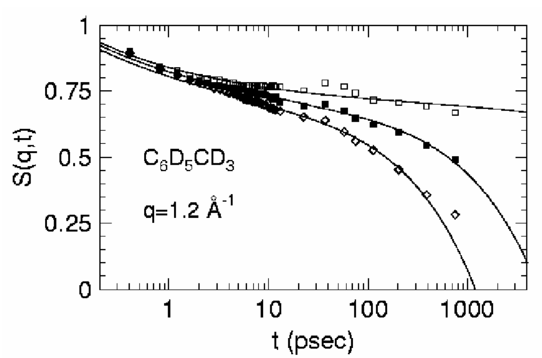
Scopigno et *al.* Science 2003

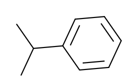


$Q = 2\mathrm{nm}^{-1}$

IXS ESRF





Cumene  ID16

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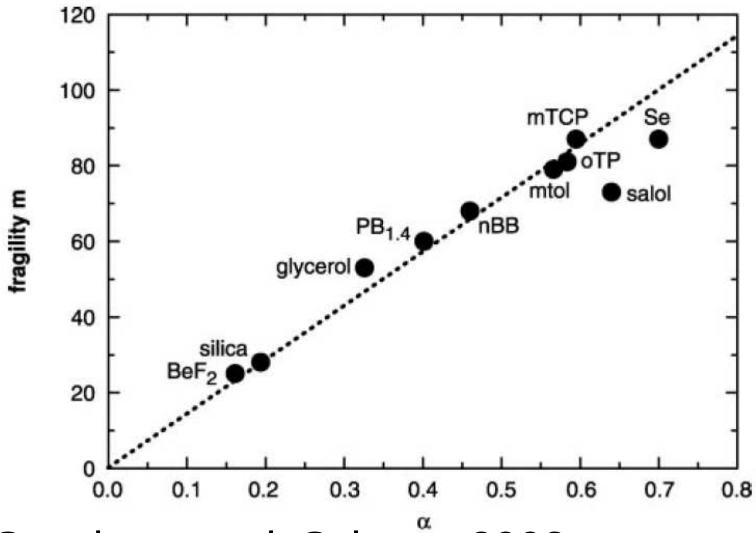
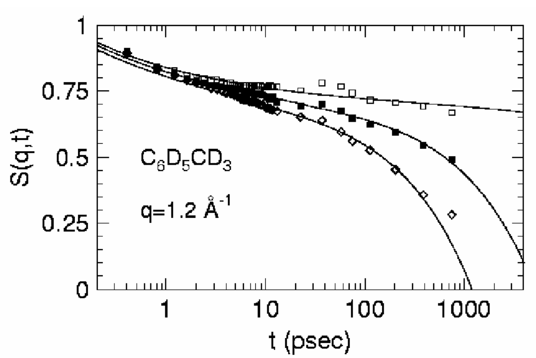
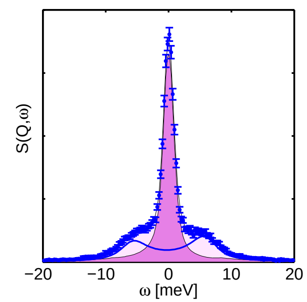


Scopigno et *al.* Science 2003





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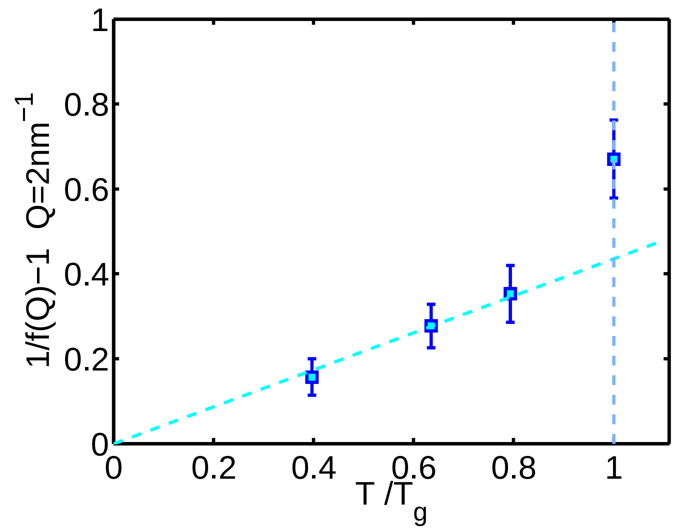
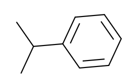
IXS ESRF



Cumene  ID16

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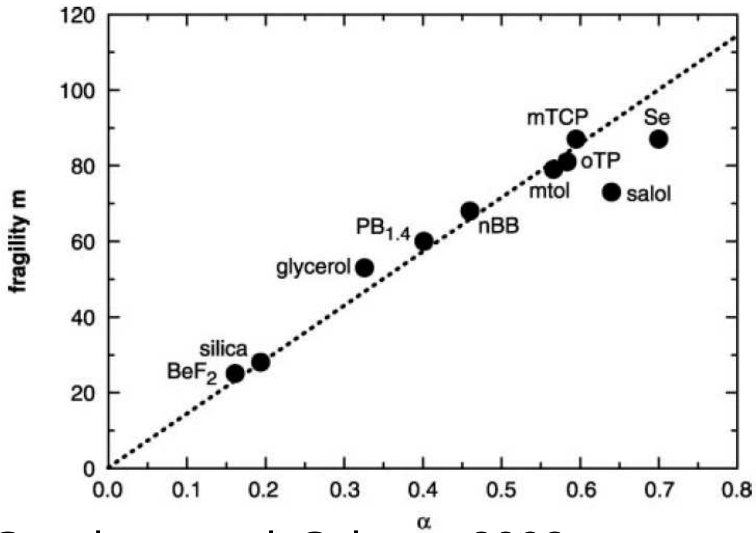
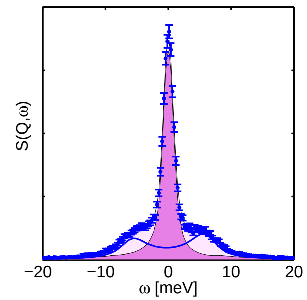
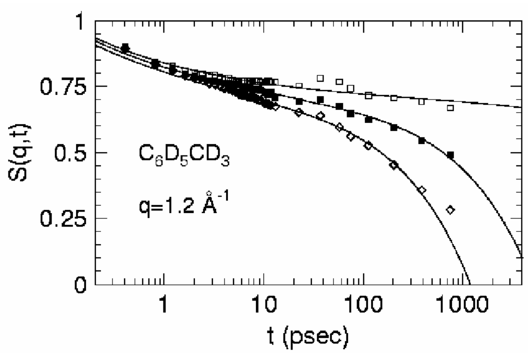


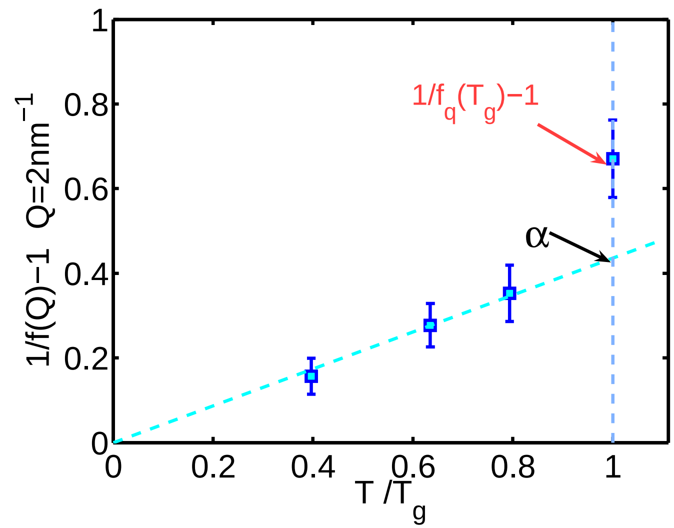
Scopigno et *al.* Science 2003

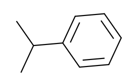


$Q = 2\mathrm{nm}^{-1}$

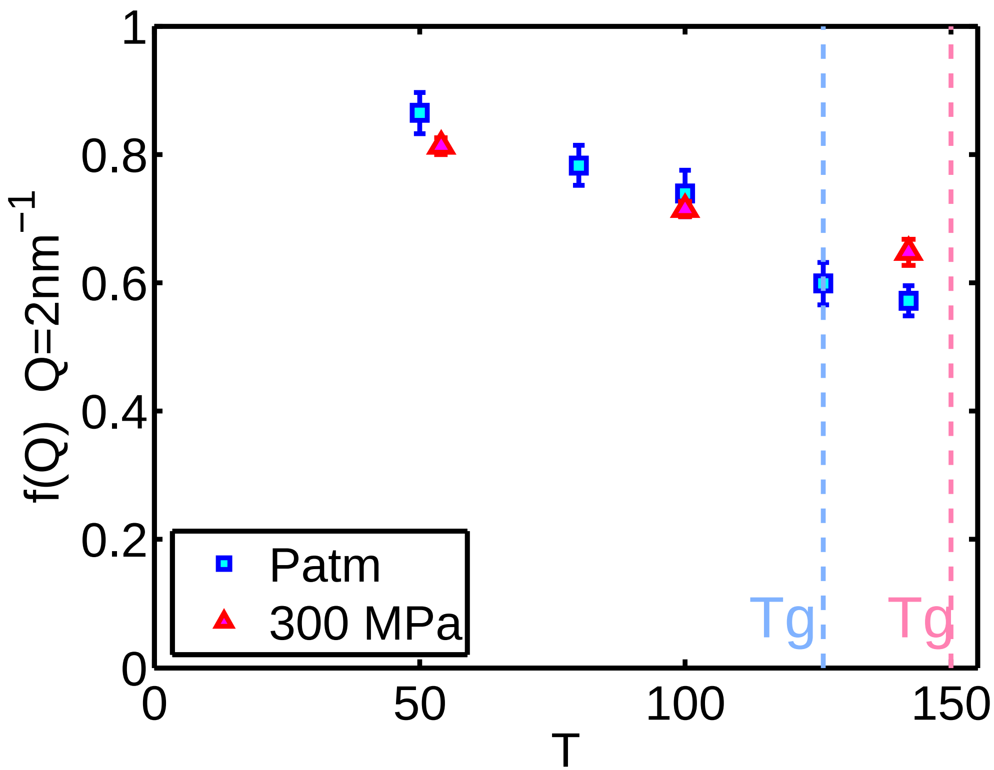
IXS ESRF

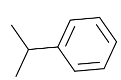




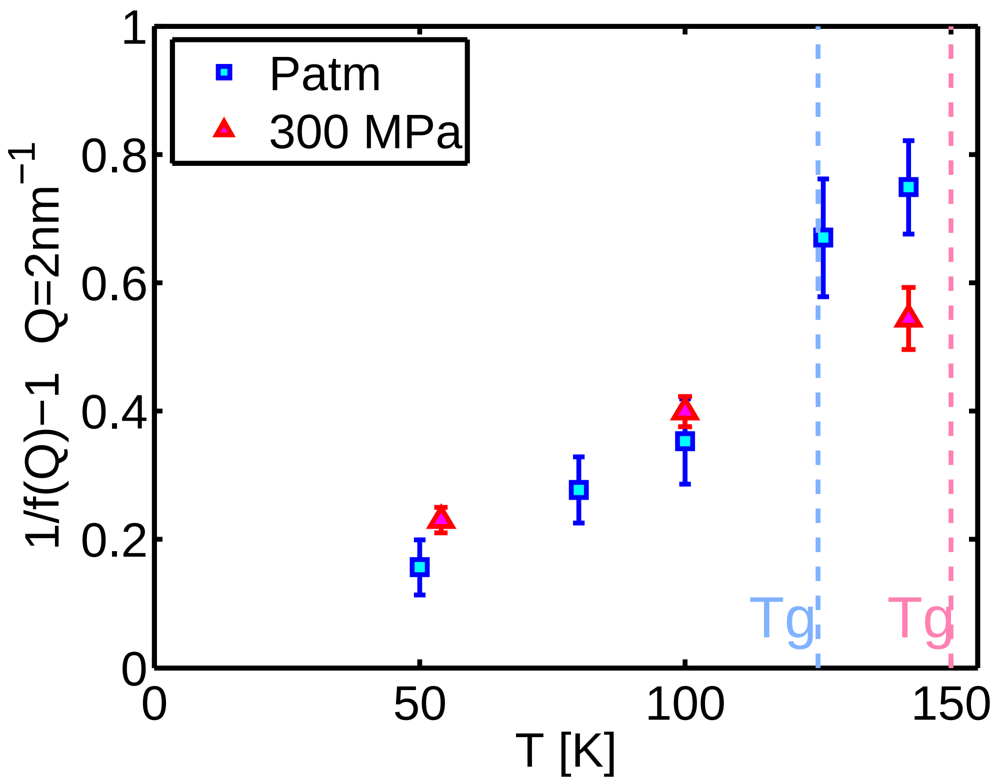
Cumene  ID16

f_q and pressure



cumene 

f_q and pressure



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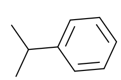
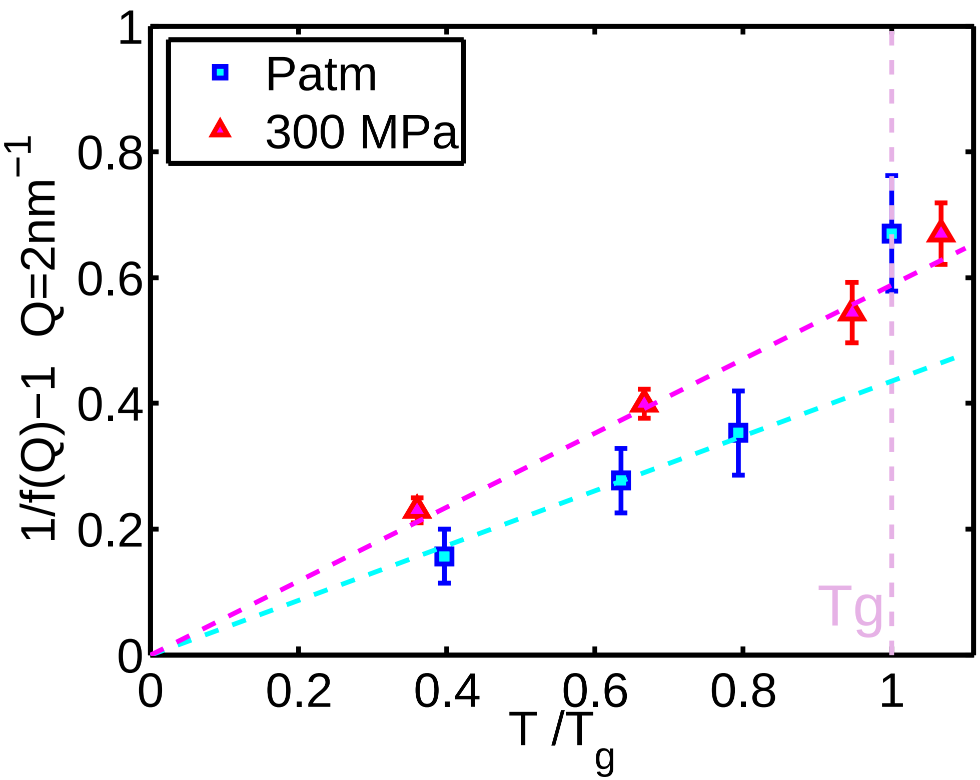
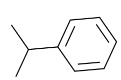
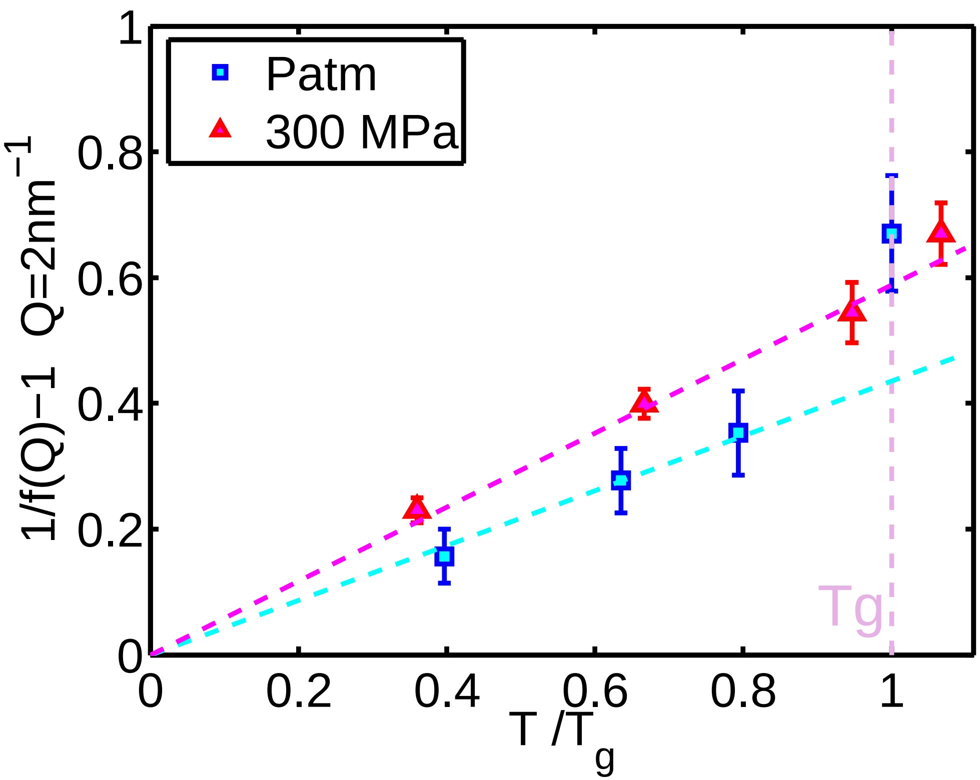


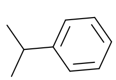
f_q and pressure



cumene 

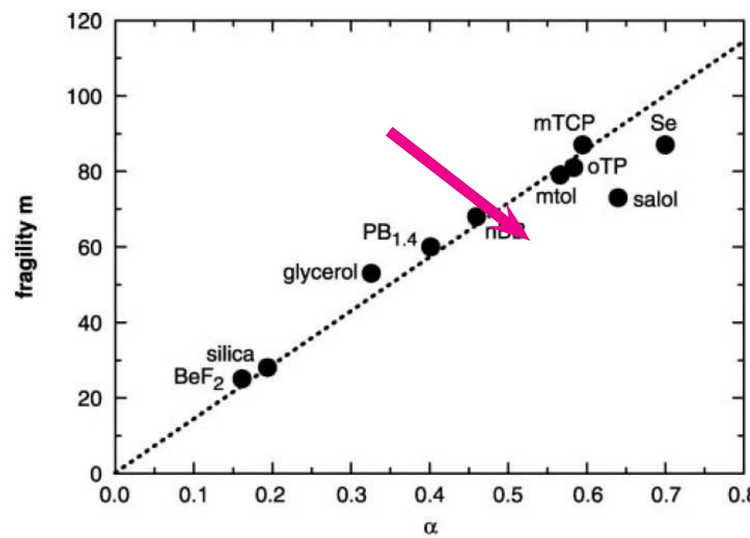
f_q and pressure



cumene 

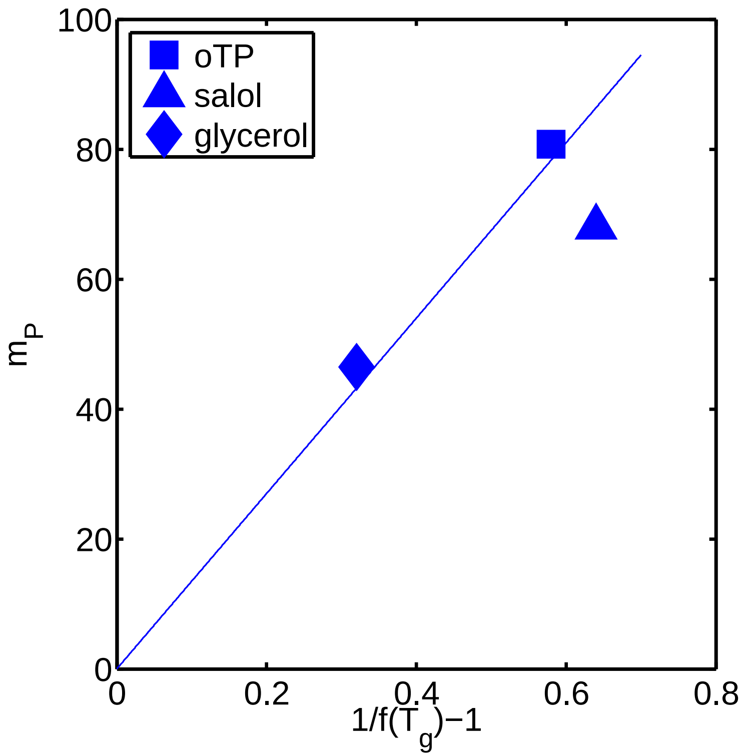
$\alpha \nearrow 33\%$

$m_P \searrow 20\%$



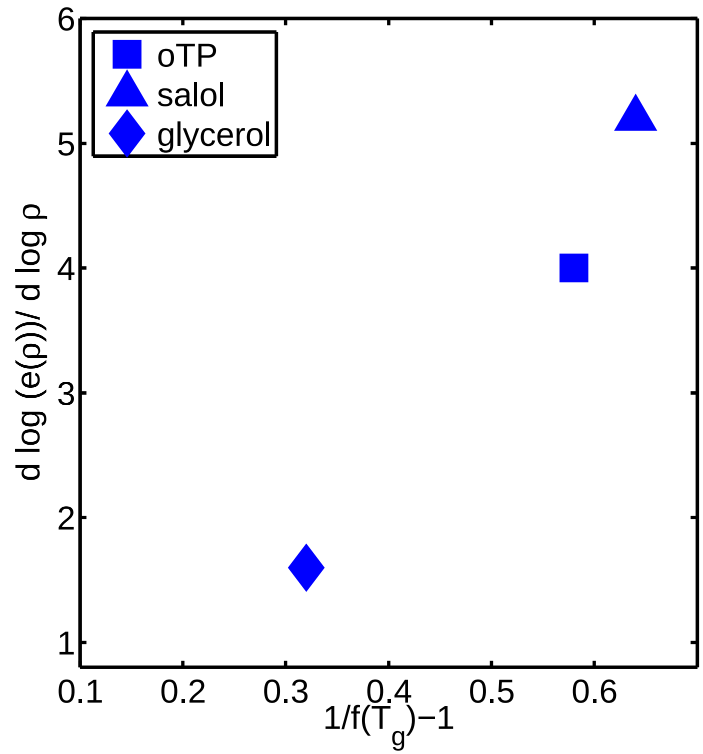
$f_q(T_g)$ **and** m

isobaric fragility



$f_q(T_g)$ and density

$$m_P = m_\rho \left(1 + \alpha_P T_g \frac{\mathrm{d}\log e(\rho)}{\mathrm{d}\log\rho}\right)$$



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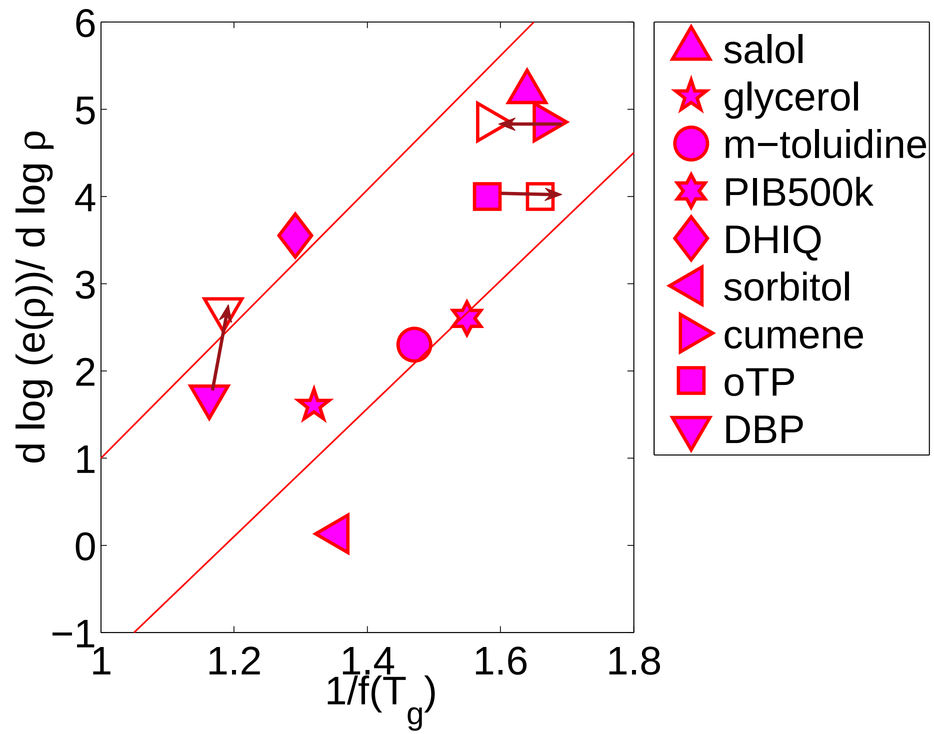
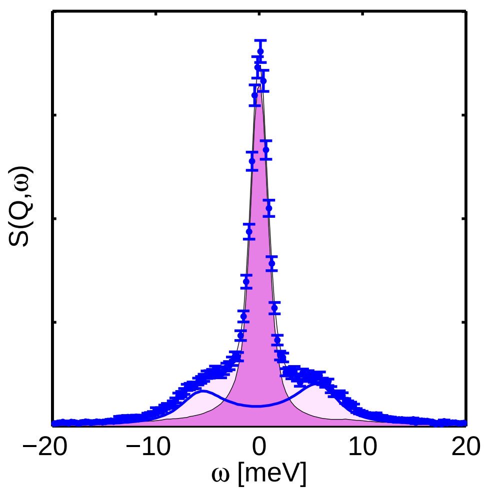
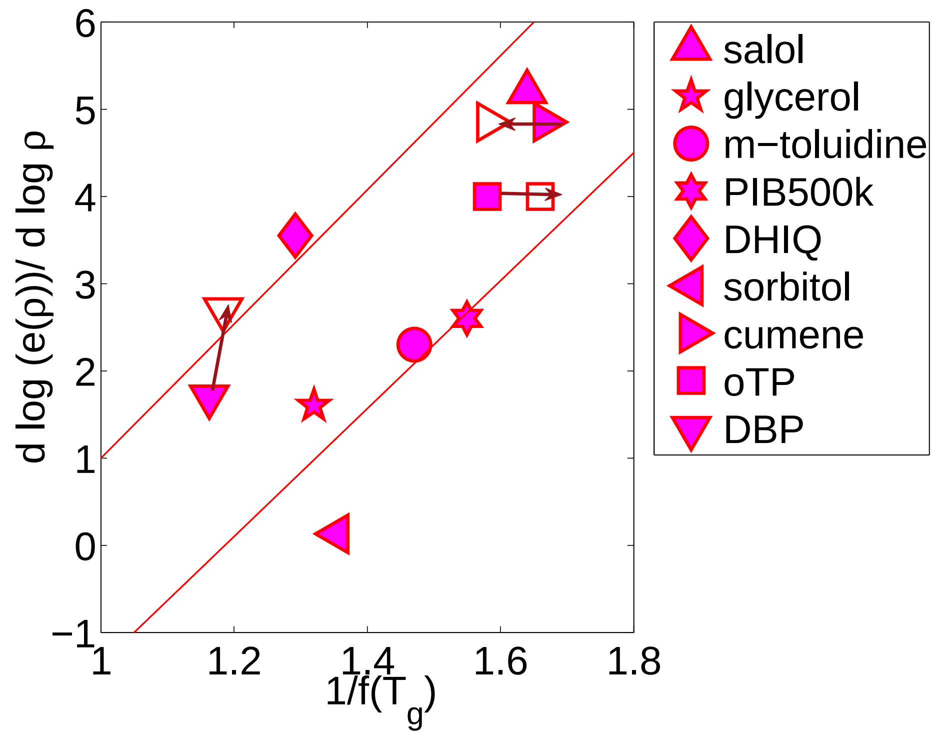


$f_q(T_g)$ and density

$$m_P = m_\rho \left(1 + \alpha_P T_g \frac{\mathrm{d}\log e(\rho)}{\mathrm{d}\log\rho}\right)$$

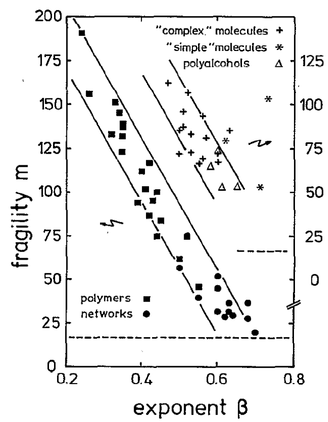
if $f_q(T_g)$ correlates to the effect of density



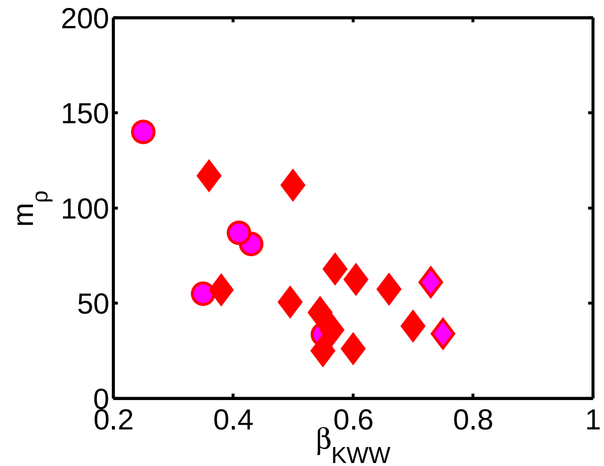


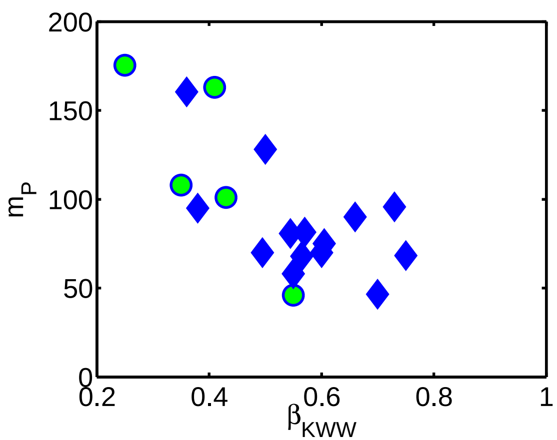
large vibrational amplitude → strong density dependence

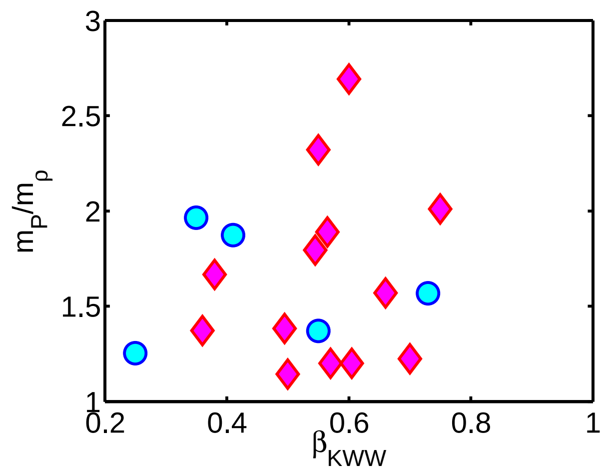
β_{KWW} and fragility



Böhmer et *al.* 1993







Niss et *al.* 2007

Conclusion

- The isobaric fragility contains information on density dependence
- Different correlations between isobaric fragility and other properties can be related to density effects, temperature effects or both of these.