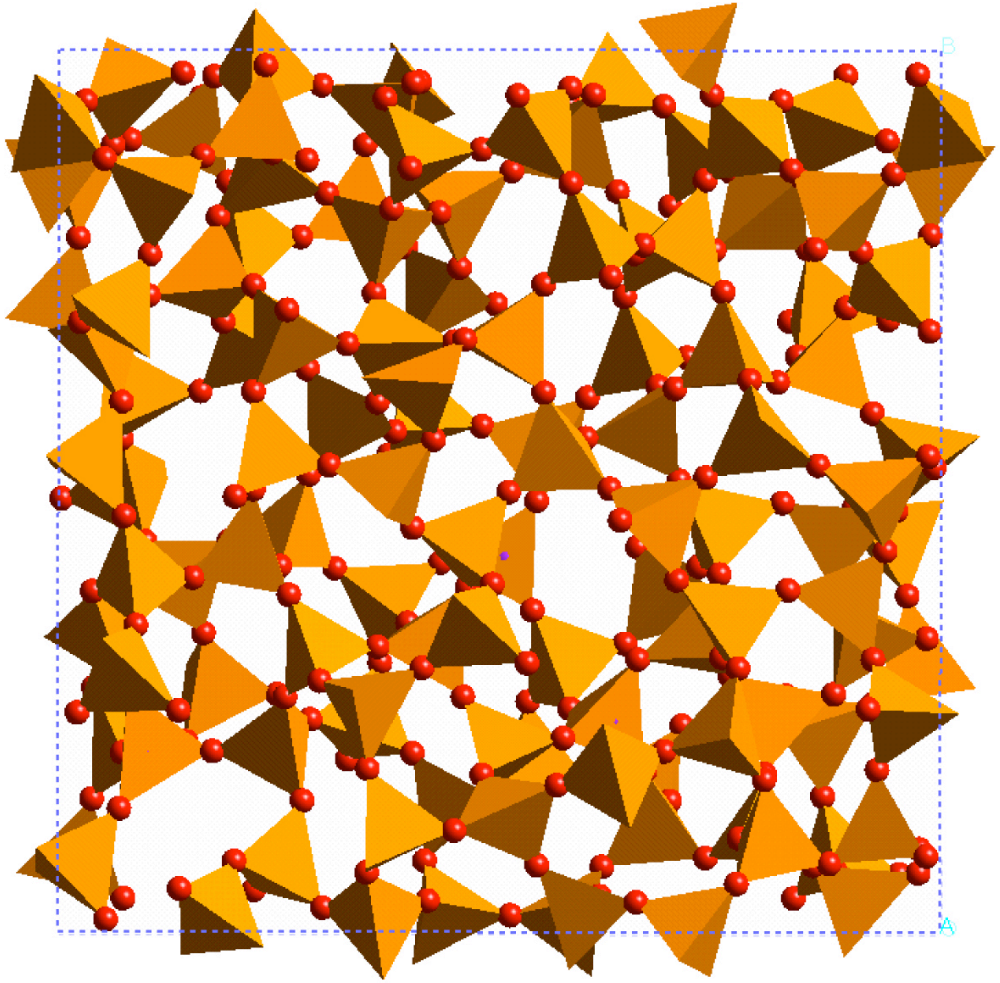
Probing the anomalous compressibility of silica glass using grid computing

Andrew M. Walker

awal05@esc.cam.ac.uk



Irreversible density change with pressure above ~25GPa

Negative thermal expansion below room temperature

Si increases co-ordination 10-25GPa (amorphous-amorphous phase transition)

Compressibility increases with pressure to ~2GPa

Low temperature anelasticity

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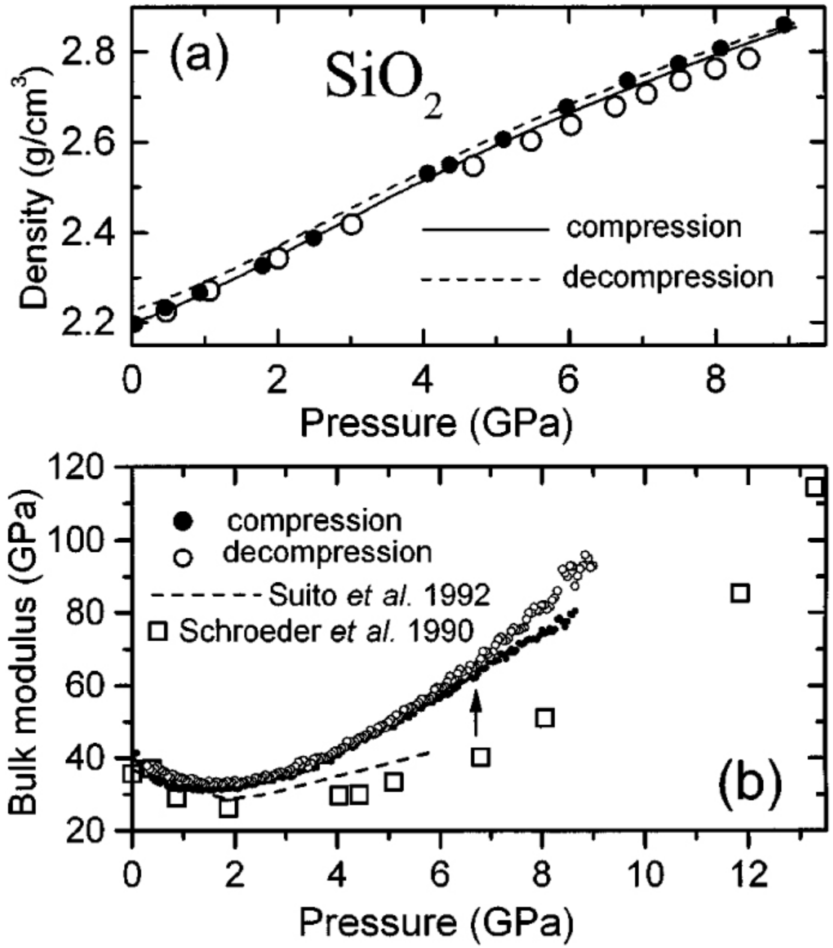
Compressibility maximum



- Density is not quite linear: the gradient is larger in the middle of the plot than at either end.
- Bulk modulus has minimum around 2 GPa: compressibility = 1/B has maximum

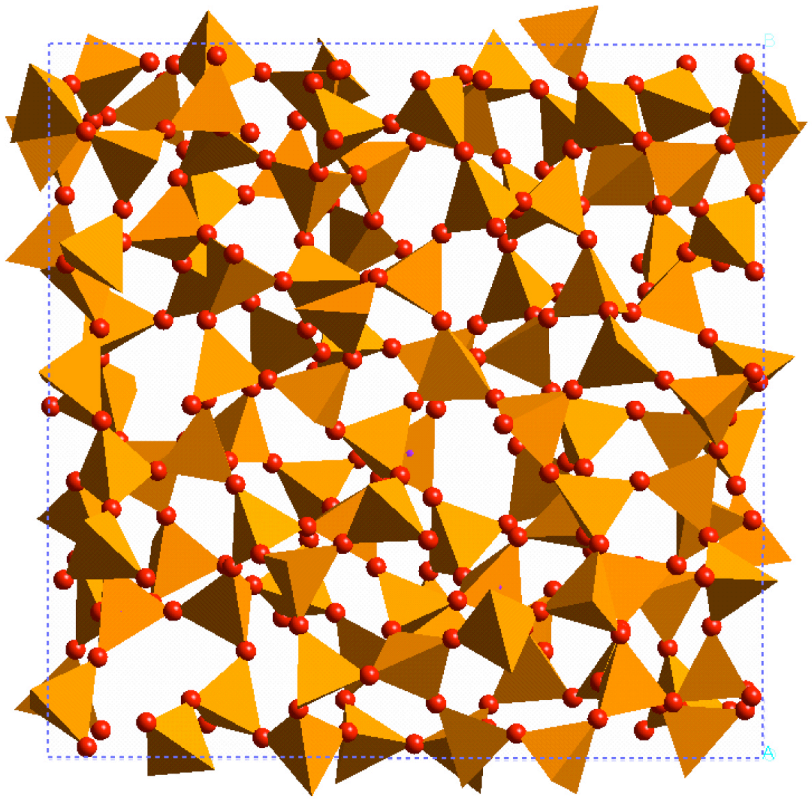
Tsiok *et al.* (1998) PRL **80** pp. 999-1002

Working hypothesis

- We expect materials to get harder under higher pressure because atoms are squashed together
- So we expect silica to be softer at lower pressures, but why will it become harder at even lower pressure?
- Point 1: in the limit of negative pressure we stretch the Si–O bonds, which costs energy
- Point 2: in between the stretched and squashed limits we believe silica glass has a lot of flexibility

Approach

- Perform molecular dynamics calculations over a range of pressures
- Examine structure and dynamics for evidence of network softening
- Use three networks with perfect connectivity (no defects) and two potential models



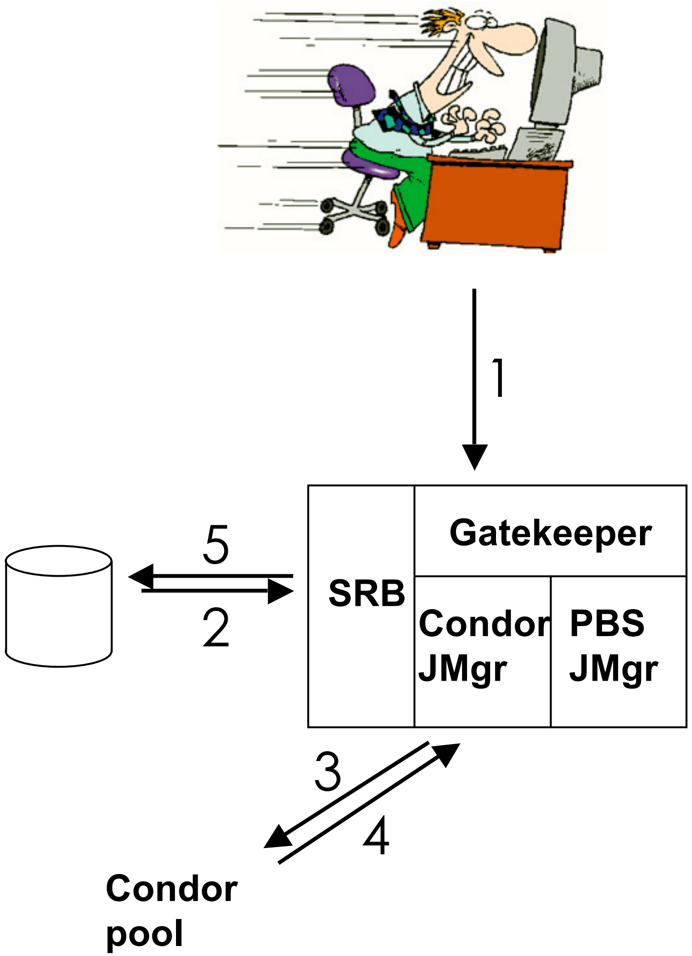
Wooten *et al.* (1985) *PRL* **54**, pp. 1392-1395

Method

- Molecular dynamics simulation with DL_POLY_3 (CML output) from -5 to 5 GPa, 0.1 GPa pressure increments
- 100 ps simulations per pressure. NPT for the first 50 ps, NVE for the second 50ps taking a snapshot of the structure every ps
- Two potential models, based on QM calculations
- Analyse configurations in terms of motion and deformation of polyhedera (GASP)
- Use distributed grid for computations and data handling

Tsuneyuki *et al.* (1988) *PRL* **61** pp. 896-872
van Beest *et al.* (1990) *PRL* **64** pp. 1955-1958

My_condor_submit

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preferredGridList = NGS, CamGrid, NWGrid
Sdir         = /home/awal05.eminerals/silica
Sget         = *
Sput         = *

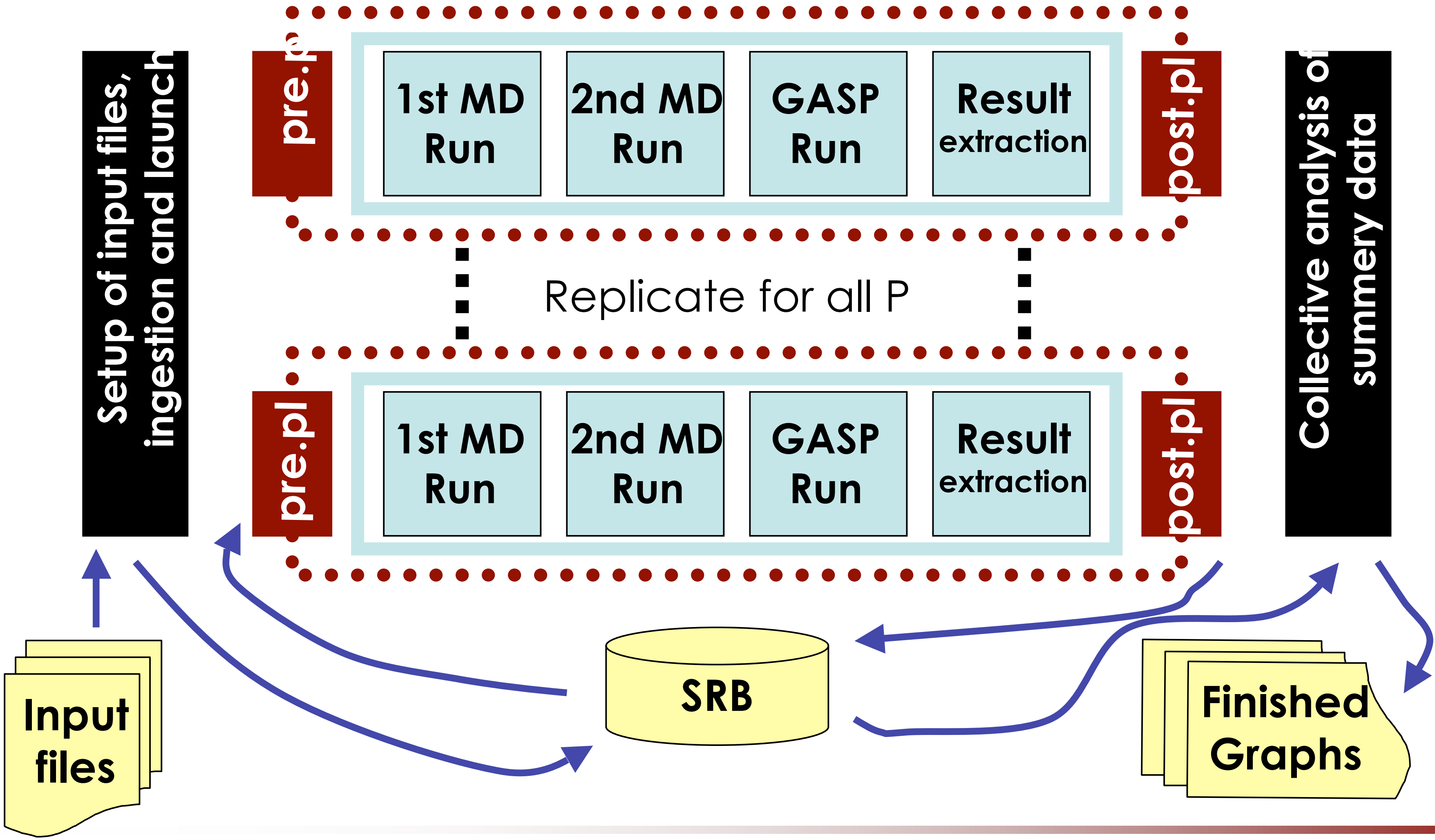
AgentXdefault = output.xml
RDesc        = Test Sanders-Catlow potential

GetEnvMetadata = true

debug = true

Queue
```

Grid computing



Automatic metadata capture

As part of the post.pl process, data is extracted from the XML output files

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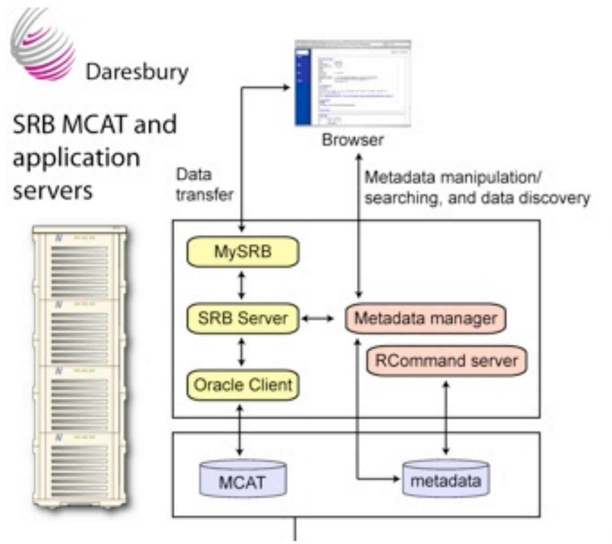
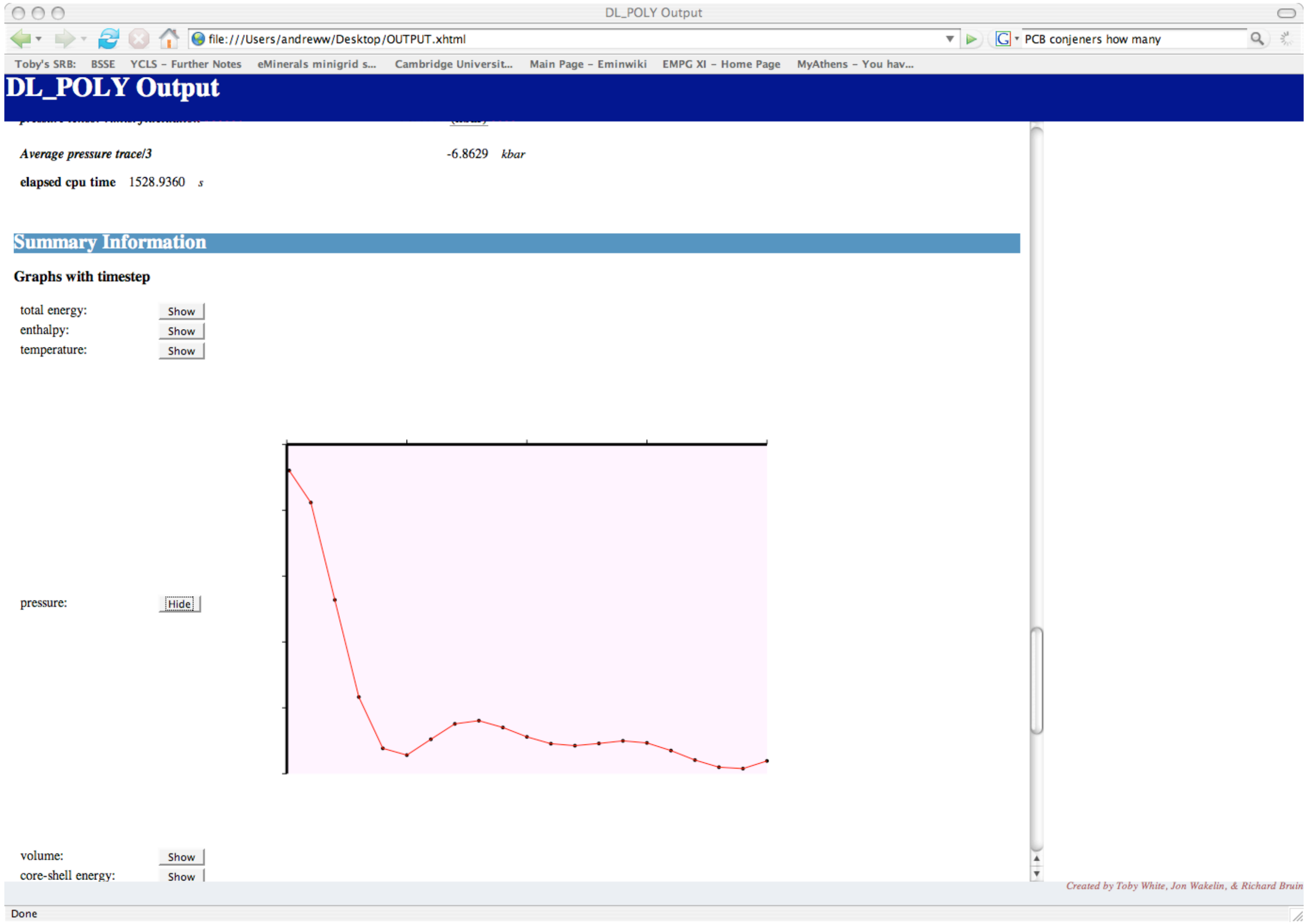
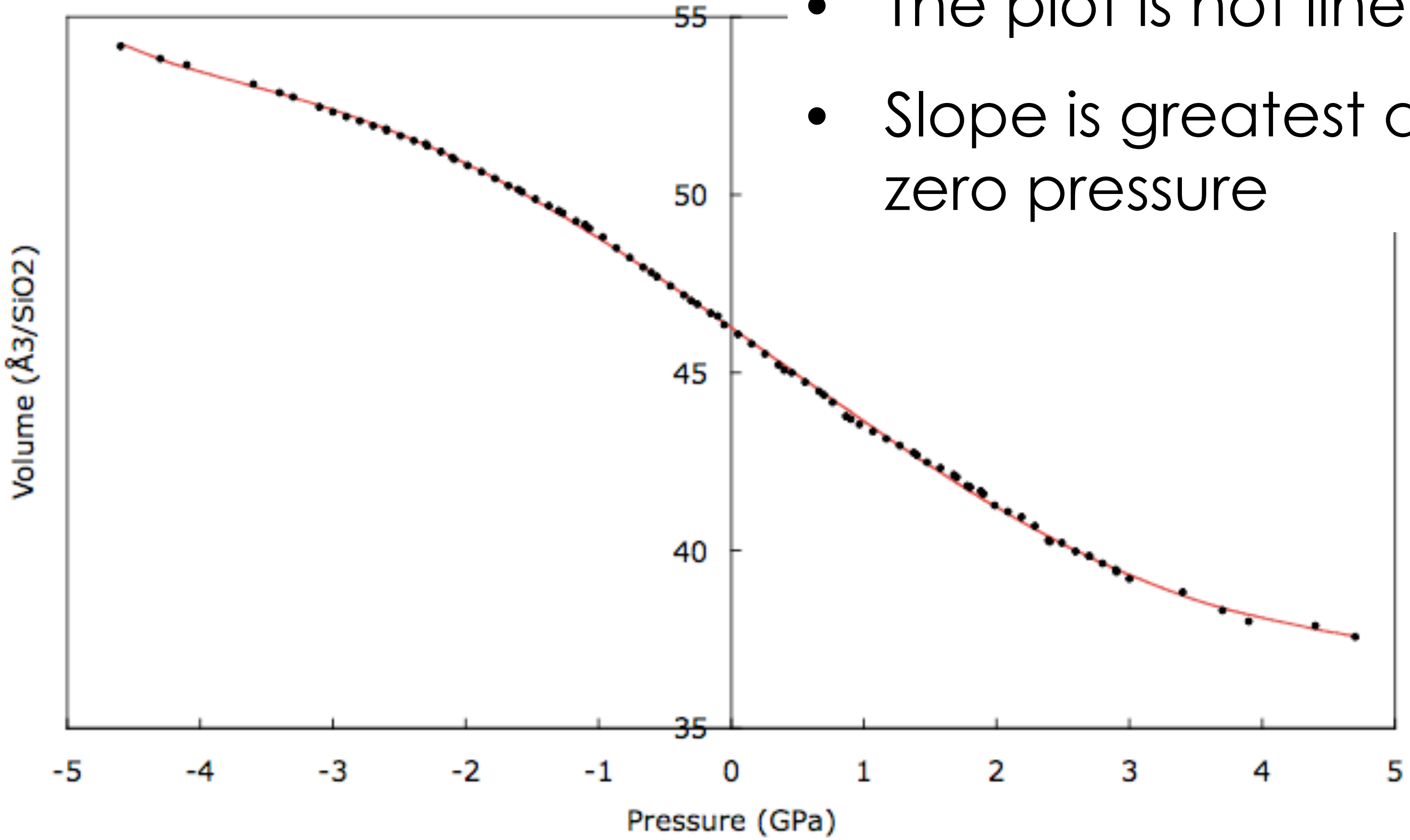


This data is added to information about the job and inserted into our metadata database and linked to the data files

Viewing results

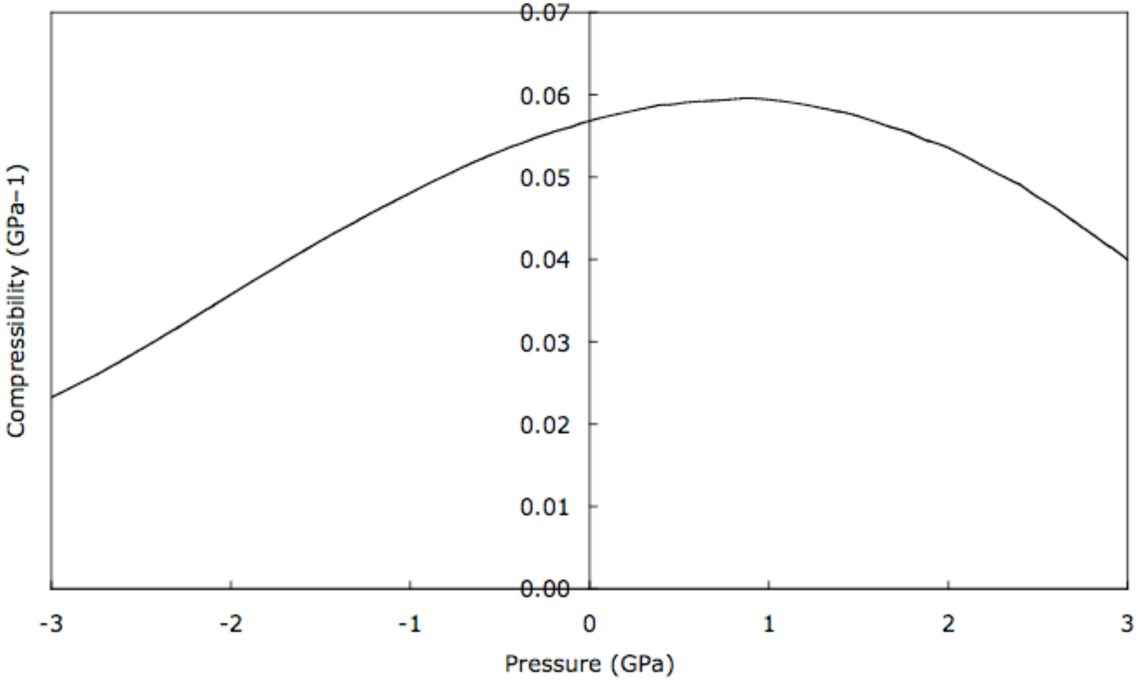


Pressure-dependence of volume



- The plot is not linear
- Slope is greatest around zero pressure

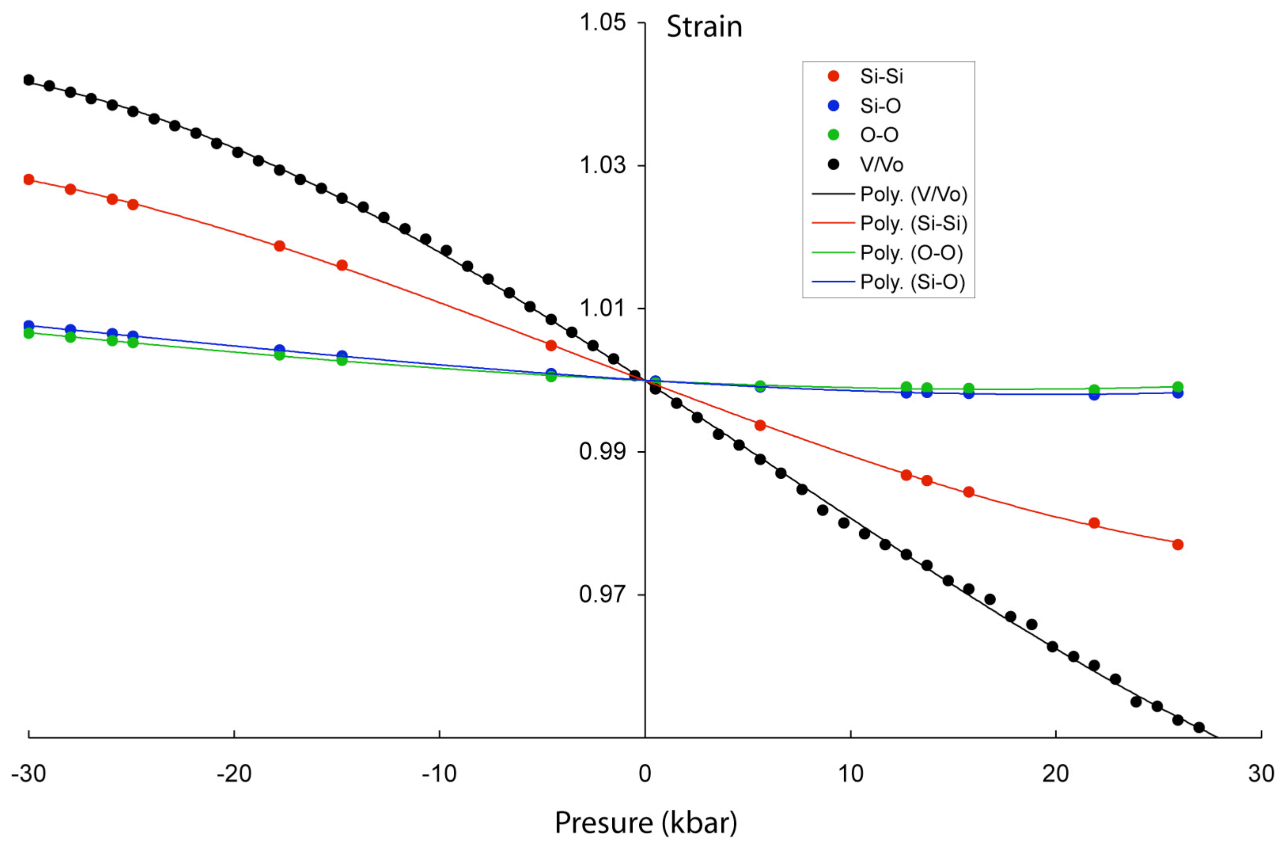
Compressibility



$$\beta = -\frac{1}{V}\left(\frac{\mathrm{d}V}{\mathrm{d}P}\right) = \frac{1}{B}$$

- Maximum around 1 GPa
- Reproduced with all potentials and networks

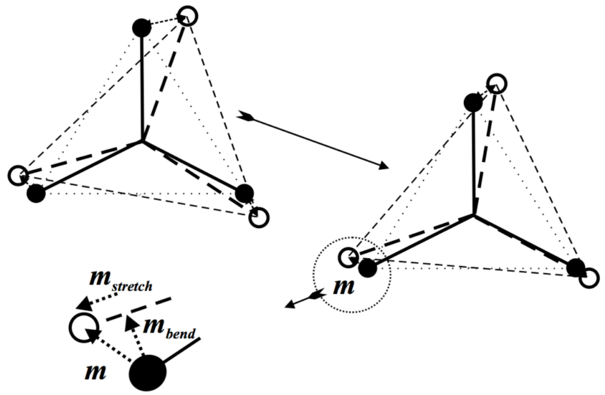
Pair distribution function



- Si–O and O–O distances do not vary much away from extremes
- Si–Si distance nearly follows sample size
- Evidence for buckling of network

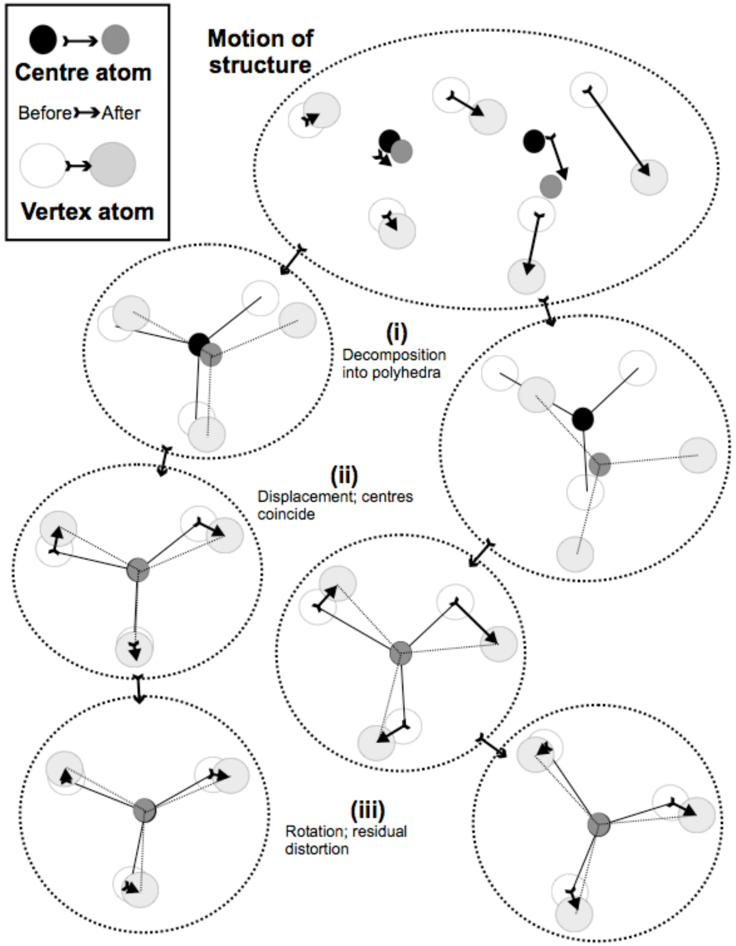
Measuring network flexibility

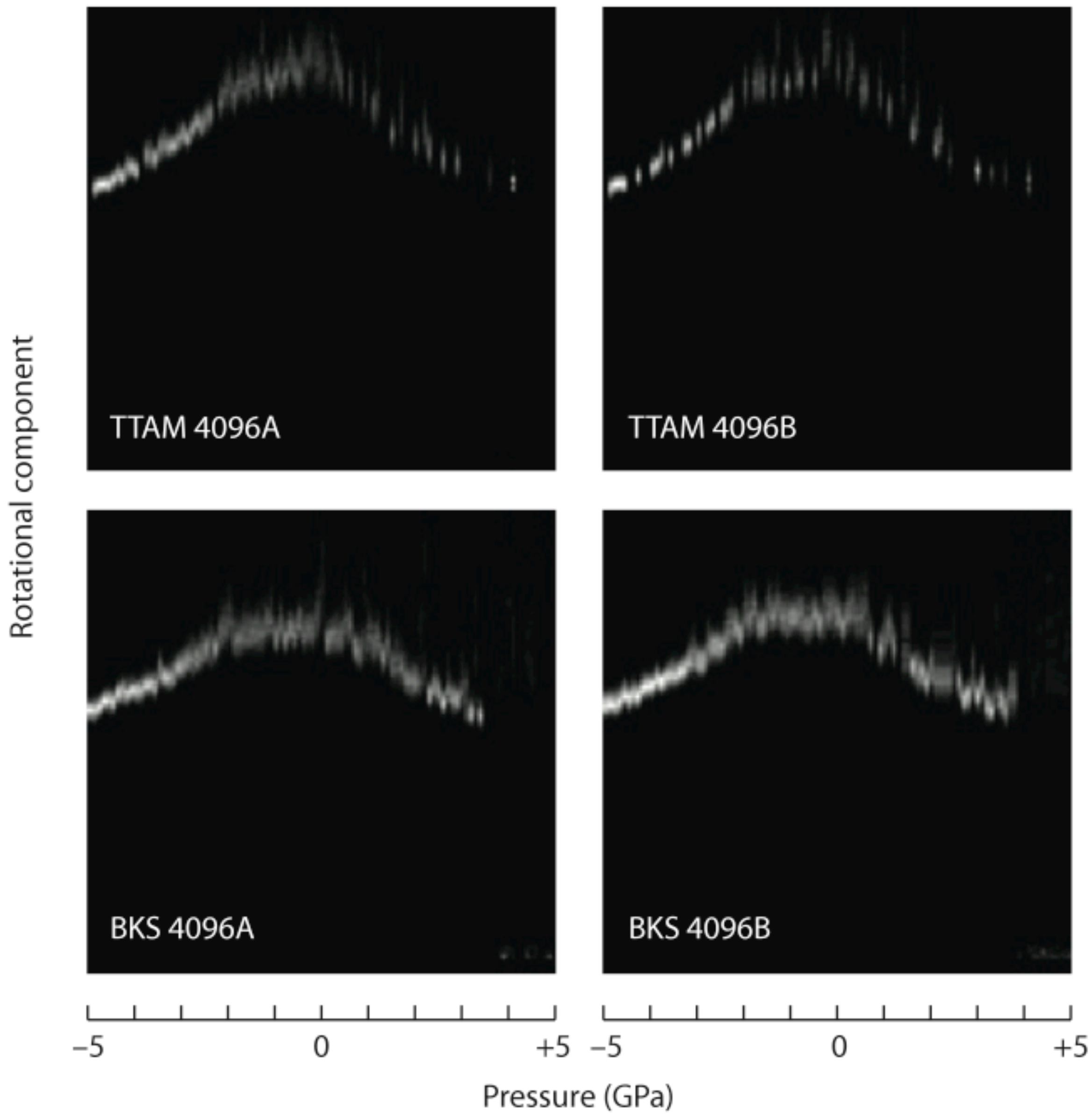
Vibration distributed into translation and rotation of tetrahedra and stretching and bending of bonds.



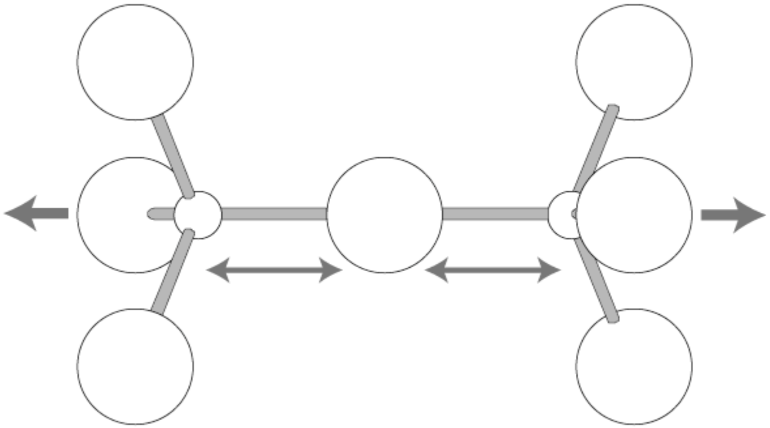
Implemented using geometrical algebra in the GASP code

Wells *et al.* (2002) *J Phys: Con Mat* **14** pp.4567-4584

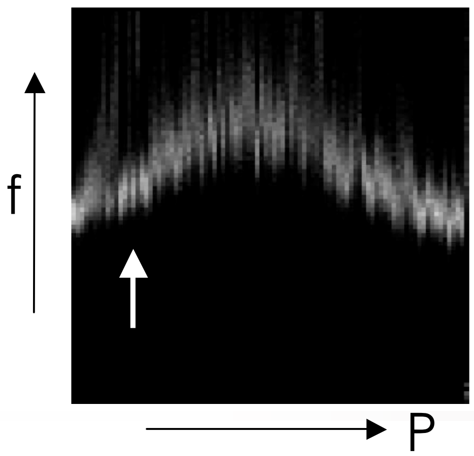



TTAM 4096A
TTAM 4096B
BKS 4096A
BKS 4096B
Rotational component
–5
0
+5
–5
0
+5
Pressure (GPa)

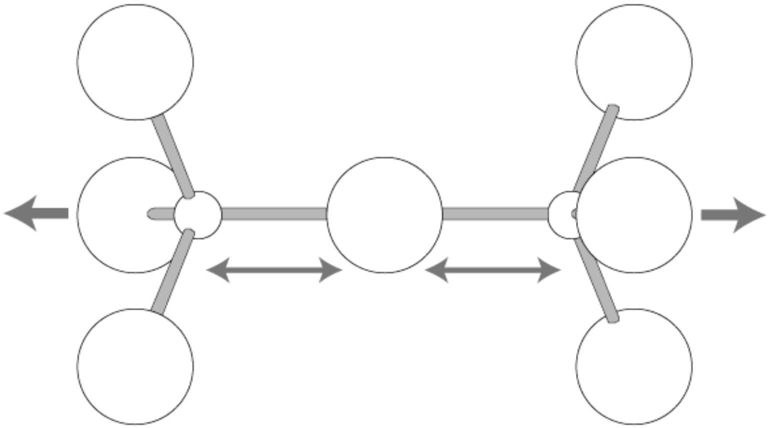
Network flexibility



Low pressure
(low flexibility)



Network flexibility



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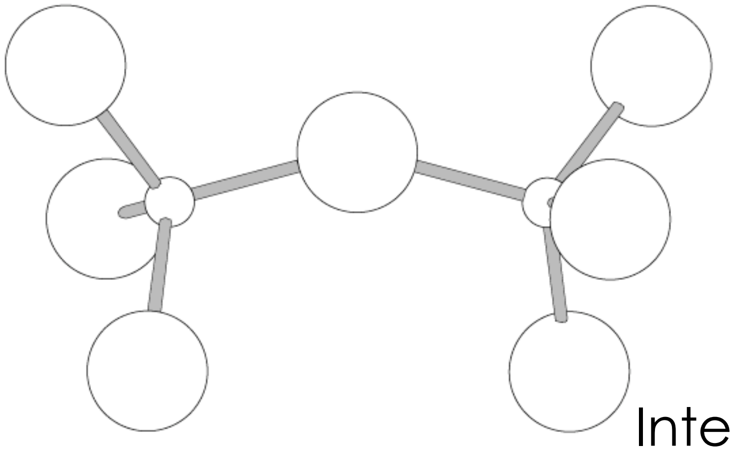
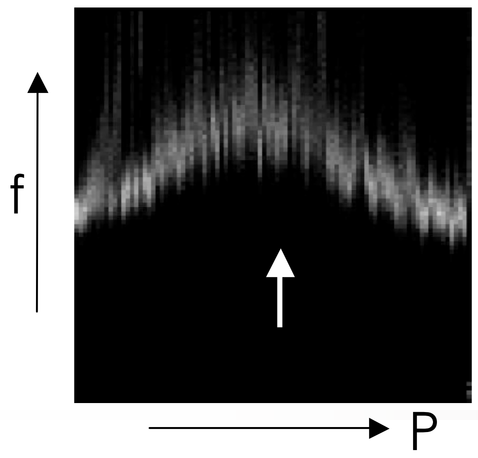
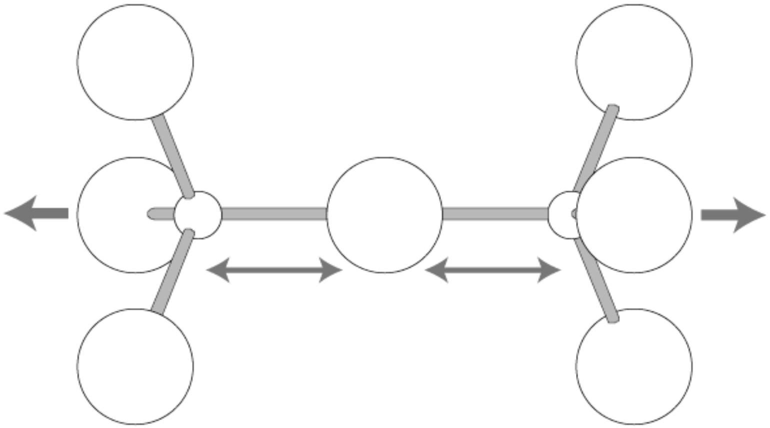


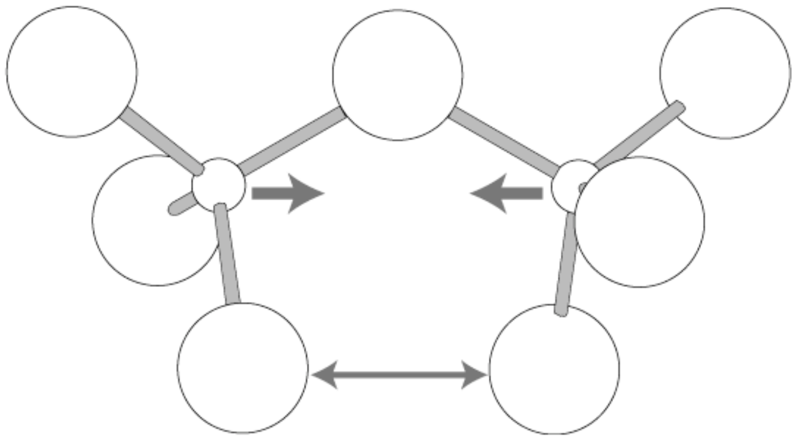
Intermediate pressure
(high flexibility)



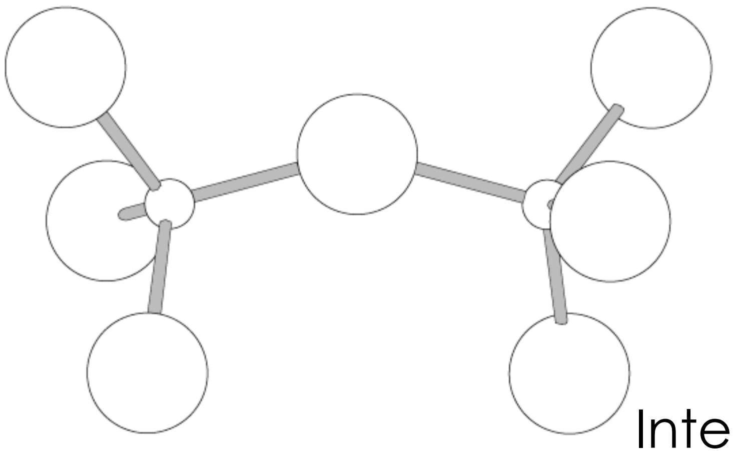
Network flexibility



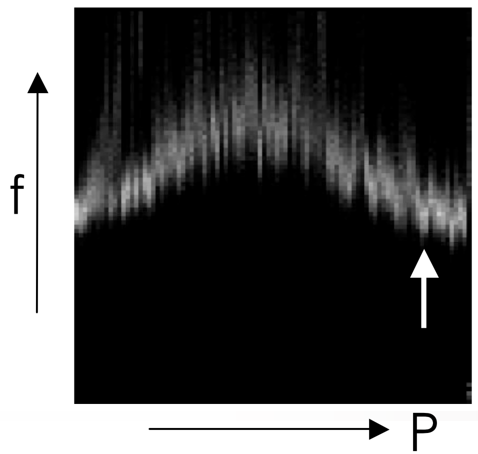
Low pressure
(low flexibility)



High pressure
(low flexibility)



Intermediate pressure
(high flexibility)



Conclusions

- The compressibility anomaly in silica glass appears to be caused by changes in the flexibility of the tetrahedral framework
- A phase transition is not required
- The mechanism does not require an amorphous state - could it be important for crystals?
- Currently investigating the anomalous thermal expansion

Walker *et al.* (2007) *J. Phys: Con. Mat.* **19** art. no. 275210
Dove *et. al.* (2006) *Mol. Simul.* **32** pp. 949-952

Acknowledgments

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