Liquid-liquid (L-L) immiscibility in supercooled aqueous solutions driven by water's polyamorphism

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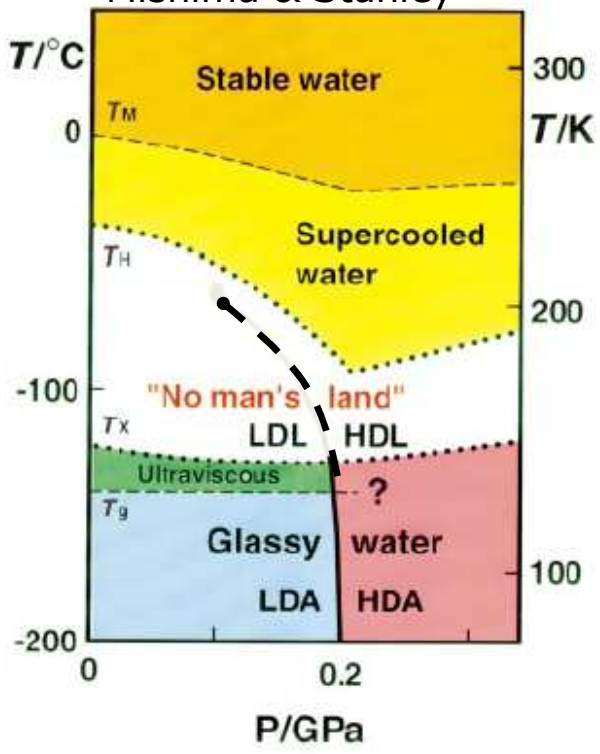


Guiding questions

1) What is the effect of hydrophilic solutes on the L-L transition?

2) Do solutes produce L-L immiscibility?

-In which concentration range?

-What are dimensions of phase segregation?

-What is composition of the phases?

3) How does topology of solutes affect L-L immiscibility ?

Molecular Dynamics Simulations

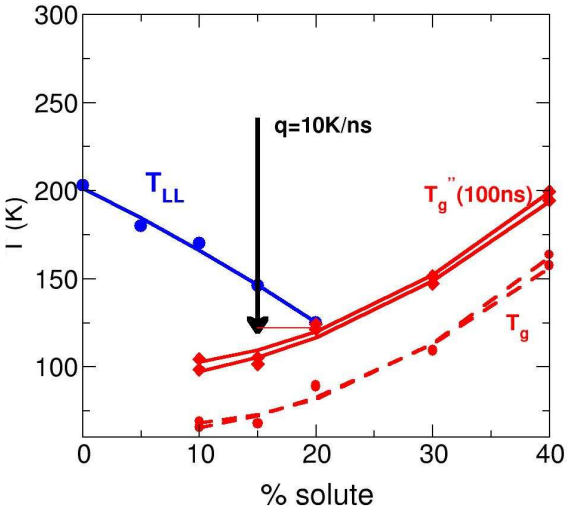
Model: Coarse grained model of water, mW (Molinero et al.) monatomic solute and homogeneous polymer with harmonic bonds.

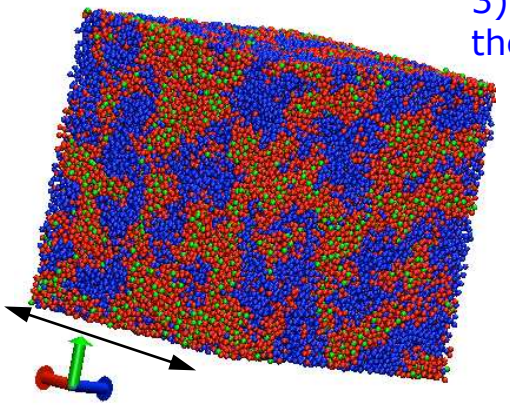
Temperature quenches, at p=0.

1) Solute concentration determines outcome on fast cooling

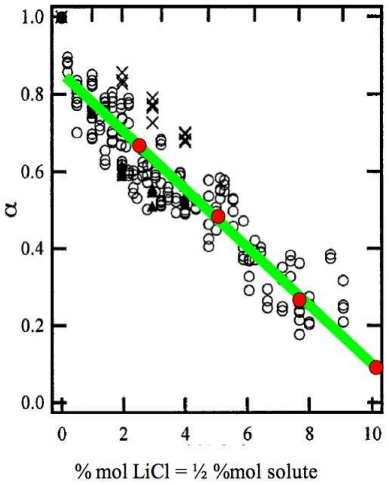
2) Hydrophilic solutes are expelled by LDL

3) Fraction of LDL in excellent agreement with the raman determination by Suzuki and Mishima





10% solute
15nm, 110592 particles
LDL: blue, HDL: red, solute: green



5) Polymeric solutes also favor L-L immiscibility

4) Size of nano-segregated phases

% solute	qmax	Size of nanodomains
5%	0.12A° $^{-1}$	5.23 nm
10%	0.15A° $^{-1}$	4.19 nm
15%	0.17A° $^{-1}$	3.69 nm