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Immense elastic softening and the responsible molecular mechanisms near the demixing phase transition of thermo-responsive polymer solutions

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Functional materials based on stimuli-responsive polymers are of great interest in view of their current use, and foreseen future implementation, in many everyday life applications. Frequent applications are sensors, drug delivery systems, soft robotics and responsive surfaces. The environmental responsiveness of such hydrogels, thin films or micellar solutions bases on a structural instability, which occurs at a demixing transition with a lower critical solution temperature (LCST).
Even though numerous theoretical and experimental studies have been carried out, central problems regarding the phase separation mechanisms remain unsolved. Using aqueous solutions of the model thermo-responsive polymer poly(N-isopropyl acrylamide) (PNIPAM), we identify the volume expansion coefficient and the isothermal compressibility as order parameter susceptibilities of the demixing transition [1-3]. An intriguing finding based on Brillouin spectroscopy is that immense strain-softening, and hence a third order elastic constant, governs the phase separation, instead of the linear elastic properties [2].
In order to better assess the relationship with the underlying molecular mechanisms, we also focus on the molecular origins of the demixing transition [1, 4]. Important variations in hydrophobic and H-bond interactions occur within the phase-separating polymer solutions. Our studies based on quasi-elastic neutron scattering show how the local diffusion behavior of the hydration water varies during the partial dehydration of the PNIPAM chains in the course of the demixing transition [4]. Novel insights into the impact of the molecular processes on the macroscopic order parameter susceptibilities of the phase separation of stimuli-responsive polymer systems are finally given.
References:
[1] M. Philipp, et al, Soft Matter, 2012, 8, 11387.
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[4] M. Philipp, et al, The Journal of Physical Chemistry B 2014, 118, 4253.

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