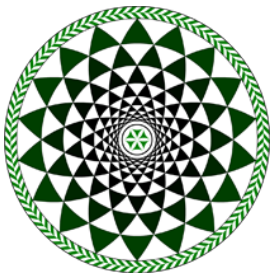

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Multiscale behaviour of confined molecular liquids and polymers

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Polyurea elastomeres exhibit phase separated mesoscopic structures with hard nanodomains embedded in a soft (compliant) matrix. This system shows (Fig.1) two glass transitions at Tg1<Tg2, which are related to the freezing of motion of molecular segments in the soft domains (α peak = usual polymer glass transition at Tg1) and to regions of restricted mobility near the hard nanodomains at Tg2 (α´-peak), respectively. We present detailed dynamic mechanical analysis (DMA) measurements [1,2] for polyureas with different molecular weight of the polyetheramines (soft domains), i.e. with varying segmental chain lengths lc and volume fractions Φx of the hard domains. It is found, that Tg1 increases drastically with increasing Φx, whereas Tg2 remains almost constant. Around Φxc≈0.25 the two curves cross, i.e. Tg1≈Tg2, which corresponds to the percolation threshold of the hard nanodomains. It means, that below Φxc the system consists of hard spheres embedded in a soft matrix and above Φxc vice versa. A very similar two glass transition behaviour was recently observed in a physically different but topologically similar system, i.e. in molecular liquids confined in nanoporous Vycor and Gelsil [3]. Here the glass transition at Tg1 occurs in the center of the nanopores whereas Tg2>Tg1 is related to the freezing of molecules near the pore walls. By functionalyzing the pore walls with silane the second glass transition at Tg2 is completely suppressed.
Following the approach of Berthier, et al. [4], we analyzed the temperature derivatives of the dynamic elastic susceptibilities to obtain the size ξ(T) of dynamically correlated regions in confined liquids and polymers. It is found, that for both systems ξ(T) increases when approaching T_g and finally diverges at TVF

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