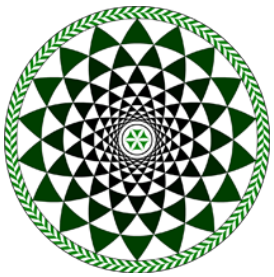

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Vapor-deposited stable glasses of glycerol: evidence for stable glass behavior and unusual transformation kinetics into the super-cooled liquid state

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Physical vapor deposition (PVD) of super-cooled liquids into the glassy state yields occasionally glasses with extraordinary high packing density resulting in an extremely low fictive temperature and high kinetic stability [1]. While a couple of organic molecules have been proven to form (ultra)stable glasses, we have recently shown that PVD might also induce enhanced molecular order in H-bonded liquids like glycerol that even persists after recovery of the liquid state on time scale up to 10¹⁰ times the structural relaxation time tau;_{alpha;} [2].
This paper focuses on the structural dynamics of ultra-thin films of glycerol prepared by organic molecular beam deposition (OMBD) at temperature well below the glass transition temperature (T_g). By using in-situ, chip-based ac-calorimetry and broadband dielectric spectroscopy (BDS), and by monitoring the sample mass and desorption phenomena by gravimetric (QCM) and pressure measurements, we were able to investigate the specific heat and relaxation dynamics of glycerol films in the glassy and liquid state systematically.
Slowly deposited films of glycerol revealed true stable glass behavior, a feature that was regarded as unlikely by other authors [3] based on dynamic fragility considerations.
Even more strikingly, we found that devitrification of glassy glycerol proceeds in a two-step scenario: The majority of the (liquid) specific heat is recovered just above the bulk glass transition temperature, while the full recovery just occurs in a second transformation that goes along with the disappearance of an excess contribution in the dielectric relaxation strength as reported in our previous work [2].
All findings and their implications are discussed in the framework of the two-order parameter model by Tanaka [4] and similar approaches.

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