

MAR15-2014-020544

Abstract for an Invited Paper
for the MAR15 Meeting of
the American Physical Society

Simple aging in molecular glasses

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The glass transition takes place when the structural (α) relaxation freezes in and the liquid enters a non-equilibrium solid state. This usually happens when the relaxation time, τ , reaches a timescale of 1000 seconds, and $\tau = 1000$ s is pragmatically used as a definition of the glass transition temperature T_g . However, if the glass is studied on a long enough time scale then relaxation is still seen as physical aging. Aging is a non-linear signature of the α relaxation in which the relaxation dynamics changes as a function of how far the system has relaxed. If the system is studied well below T_g then equilibrium will not be achieved, but just below or around T_g it is possible to systematically monitor the non-linear relaxation all the way to equilibrium. We have developed a micro cryostat which is optimized for making fast changes in temperature and keeping temperature stable over days and even weeks. Combining this micro cryostat with a small dielectric cell it is possible to monitor non-linear relaxation in a dynamical range of more than 4 decades from 10 seconds to a 10^5 seconds. The aging is monitored after a fast temperature jump. This means that the aging itself is isotherm, and the data therefore directly shows, how the relaxation-rate changes as volume and structure change on the isotherm. We have studied several molecular liquids and find that the data to a very large extend can be understood in terms of a TNM formalism. This implies time-aging-time superposition and suggests a simple picture where the out of equilibrium “states” correspond to equilibrium states - at an other temperature. If the α relaxation is dynamically heterogeneous as it is commonly believed, then the aging results show that fast and slow “modes” of the relaxation are governed in the same way by structure and volume. We hypothesize that aging according to TNM formalism is an intrinsic property of Roskilde Simple liquids.