Abstract Submitted
for the MAR15 Meeting of
The American Physical Society

Dynamic Correlation Length Scales under Isochronal Conditions at High Pressures RICCARDO CASALINI, Naval Research Laboratory — The origin of the dramatic changes in the behavior of liquids as they approach their vitreous state – increases of many orders of magnitude in transport properties and dynamic time scales – is a major unsolved problem in condensed matter. These changes are accompanied by greater dynamic heterogeneity, which refers to both spatial variation and spatial correlation of molecular mobilities. The question is whether the changing dynamics is coupled to this heterogeneity; that is, does the latter cause the former? To address this we carried out the first nonlinear dielectric experiments at elevated hydrostatic pressures on two liquids, to measure the third-order harmonic component of their susceptibilities. We extract from this the number of dynamically correlated molecules for various state points, and find that the dynamic correlation volume for non-associated liquids depends primarily on the relaxation time, sensibly independent of temperature and pressure. We support this result by molecular dynamic simulations showing that the maximum in the four-point dynamic susceptibility of density fluctuations varies less than 10% for molecules that do not form hydrogen bonds. Our findings are consistent with dynamic heterogeneity serving as the principal control parameter for the slowing down of molecular motions in supercooled materials.

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Date submitted: 03 Dec 2014

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