Ultra-long-range dynamic correlations during aging of gels

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Gels are ubiquitous in our daily life and understanding the mechanisms via which they form remains a challenging task, specially in the context of designing such materials with specific mechanical response. A common method to form gels is via a nonequilibrium route, during which a homogeneous fluid is thermally quenched into a phase coexistence region. Such a quench process generates bicontinuous structures and when the dense phase forms an amorphous solid, the phase separation is kinetically hindered leading to the formation of a gel.

The aging dynamics during the formation of such a non-equilibrium gel has been investigated in diverse experiments and the observations differ qualitatively from those in liquids and glasses. The most striking feature is the finding that time correlation functions are described by compressed exponential relaxations, to which has also been associated the existence of long-ranged spatial correlations in the dynamics. A miscroscopic understanding of such *anomalous* behaviour is still missing.

Using large-scale computer simulations, we explore the aging dynamics in a microscopic model for gels, following a thermal quench into the phase coexistence region. We find that gelation resulting via an arrested phase separation process is accompanied by the *anomalous* dynamical signatures as reported in experiments, viz. occurrence of superdiffusive particle motion and correspondingly compressed exponential relaxation of time correlation functions (left panel of Fig.1). Further, via a spatio-temporal analysis of the dynamics, we demonstrate that intermittent heterogeneities produce spatial correlations over extremely large length scales; see right panel of Fig.1.

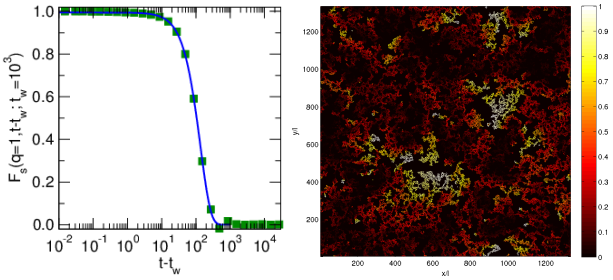

FIG. 1. (Left) Self-intermediate scattering function, measured for wave-vector $q = 1$, corresponding to an age $t_w = 10^3$ of the gel, following a thermal quench. The straight line shows that the data can be fitted with a compressed exponential. (Right) Spatial map of mobility, during the aging process relative to the same age, demonstrating that the size of mobile/immobile domains extends over large length-scales.

Thus, our study provides a microscopic insight into the spontaneous aging processes observed experimentally in gels and other similar soft materials.

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[1] P. Chaudhuri, and L. Berthier, *arXiv:1605.09770.*