Aging, stiffening and softening of gel networks

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Disordered elastic solids of soft condensed matter like proteins, colloids or polymers are ubiquitous in nature and important for modern technologies. They can form even at very low solid volume fraction via aggregation into a variety of complex and often poorly connected gel networks [1, 2]. In most cases, the interaction energies and the size of the aggregating units make these structures quite sensitive to thermal fluctuations, with a rich relaxation dynamics [3–14], associated to spontaneous local reorganizations often referred to as "micro-collapses". Investigating how such different dynamical processes emerge at rest and how they depend on the material microstructure remains a challenge.

We have used 3D numerical simulations of model solids [15] to show that, the relaxation dynamics underlying the aging change dramatically if enthalpic stresses, frozen-in upon solidification are significantly larger than Brownian stresses [16]. The timescales governing stress relaxation respectively through thermal fluctuations and elastic recovery are key: when thermal fluctuations are weak with respect to enthalpic stress heterogeneities, the stress can partially relax through elastically driven fluctuations. Such fluctuations are intermittent, because of strong spatio-temporal correlations that persist well beyond the timescale of experiments or simulations, and the elasticity built into the solid structure controls microscopic displacements, leading to the faster than exponential dynamics reported in experiments and hypothesized by recent theories [17]. Thermal fluctuations, instead, disrupt the spatial distributions of local stresses and their persistence in time, favoring a gradual loss of correlations and a slow evolution of the material properties.

In addition to affecting the time evolution of the material properties at rest, these processes interplay with an imposed mechanical load or deformation [18–22] and hence may be crucial for the mechanical response of this class of solids. We show how tuning the structural connectivity and the local internal stresses controls the non-linear mechanical response under shear deformations. Our model gels exhibit strong localization of tensile stresses that may be released through the breaking of bonds, leading to a strain softening and/or strain hardening. Our findings help to rationalize the non-linear behavior highlighted in various experimental observations [2, 23].

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