

Glass is a matter of time

Daniele Coslovich

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Understanding the behaviour of viscous liquids close to the glass transition is a century-old problem. The microscopic underpinnings of their mechanical response have now been made clearer by a unified percolation description, in both two and three dimensions.

Liquids normally flow rapidly when subject to a mechanical perturbation. As they are cooled towards the glass transition, however, flow occurs on extraordinarily long time scales, irrespective of molecular details and chemical composition. At the atomic scale, such slow dynamics are governed by a complex hierarchy of relaxation processes that are not easy to reconcile into a coherent picture. Writing in *Nature Physics*, Liang Gao and colleagues¹ rationalize the mechanical response of a range of metallic glass-formers within a double percolation scenario, suggesting a simple and unified description of glassy dynamics.

To understand percolation in its simplest form, consider a square lattice whose sites can be either free or occupied. If we increase the fraction of occupied sites, we eventually reach a critical threshold at which a connected group of occupied sites percolates: it spans the system across one of its dimensions. The statistical physics of percolation has been studied extensively in the last century², not only when sites are occupied randomly but also in the presence of correlated disorder, as well as on complex networks.

Percolation ideas have surfaced several times in the description of amorphous materials. Still, they do not feature prominently in the main theories of the glass transition, which revolve around the temperature dependence of the slow structural relaxation³. Glass-forming liquids like polymer melts, molecular liquids and metallic alloys display a wide spectrum of relaxation times, featuring often (but not always!) an additional fast process, historically called β - or Johari–Goldstein relaxation. Its signature is a distinct peak, or shoulder, in the frequency-dependent response function of the material.

The complex shape of glassy relaxation spectra has animated a decades-long debate⁴. How universal is the distinction between fast and slow processes? How do their time scales change with temperature? Gao and colleagues provide a fresh look on these issues by simulating the mechanical response of realistic models of metallic glasses. The computational protocol mimics the dynamic mechanical spectroscopy technique used in experiments: the system is subject to periodic shear at some fixed frequency and the mechanical response is determined from the oscillation of the shear stress. Figure 1a shows a typical mechanical response with well-separated fast and slow processes.

To connect the mechanical response to the atomic level, Gao and colleagues analysed the particles' motion over a time interval inverse to the deformation frequency. The particles were then partitioned into fast and slow ones according to whether they moved more than

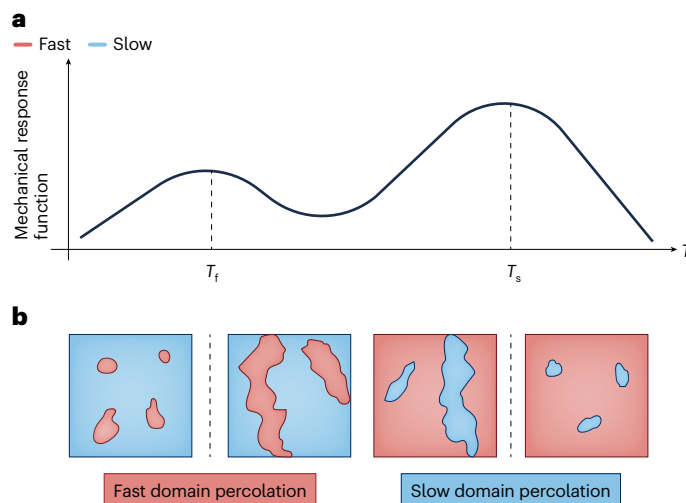


Fig. 1 | Double percolation scenario. **a**, Sketch of the mechanical response as a function of temperature T in a glass-forming liquid. The periodic shear deformation frequency is fixed. The well-separated peaks at temperatures T_f and T_s pinpoint the fast and slow processes, respectively. **b**, Percolation of slow (blue) and fast (red) regions, when displacements are measured over a time interval inverse to the frequency. The percolation temperatures occur close to the peaks of the mechanical response function.

the typical vibrational excursions: clearly, the fraction of fast particles decreases upon cooling, while the fraction of slow particles does the opposite, see Fig. 1b.

This is where double percolation kicks in: upon cooling, the slow regions percolate at a temperature that is higher than the percolation temperature of the fast regions. The key finding, highlighted in Fig. 1b, is that the two percolation temperatures closely track the peaks in the mechanical response function, no matter the probe frequency. When the percolation temperatures are too close, the fast process reduces to a wing of the slow peak. This provides a microscopic underpinning to the response spectra observed experimentally⁴.

How should one interpret the presence of two percolation transitions? If fast and slow particles were drawn at random, their percolation thresholds would be related by symmetry, as the fractions sum to unity. The twist is that the motion of particles in glass-forming liquids is strongly correlated in space and time: following a mechanical perturbation, groups of neighbouring particles start flowing locally, while others remain temporarily stuck around local equilibrium positions. In the steady state, we get a dynamic picture of fast and slow regions whose roles change constantly – this is dynamic heterogeneity, a hallmark of glassy dynamics³.

An appealing picture is that, around the glass transition, the fast regions relax within a percolating and nearly frozen matrix, in analogy to systems with quenched disorder, like fluids in porous media. The correlated motion within the fast regions is likely characterized by

dynamic facilitation, as shown by recent computational studies close to the glass transition⁵. Percolation concepts may then help to bridge the gap between dynamic facilitation and thermodynamic theories of glassy dynamics⁶.

Interestingly, when liquids are confined on a plane, no separate fast and slow processes are observed. The authors attribute this effect to a tight geometrical coupling between the percolation thresholds in 2D. One may expect that the separation of time scales between the mobilities of the chemical species also plays a role. Investigating this point will deepen our understanding of the nature of glassy dynamics across spatial dimensions⁷.

Thermally activated relaxation over large energy barriers is a common paradigm to account for slow dynamics in molecular systems. The authors invoke it to interpret their findings, suggesting that fast and slow processes, when clearly separated, are associated with small and large apparent activation barriers, respectively. To sharpen up this interpretation, however, we need a precise operational definition of energy barriers.

The potential energy landscape of glass-forming liquids is rugged and high-dimensional. Its complex hierarchical structure was explored by Heuer and co-workers in a computational tour de force during the early 2000s⁸. Recently, Pica Ciamarra et al.⁹ introduced a simpler strategy to find the barriers of individual relaxation events, as well as the activation free energy for structural relaxation. With these tools at hand, the time is ripe to clarify thermal activation in glasses, for good.

Gao and colleagues have tested the double percolation scenario on a range of metallic alloys. The generality of their observations should now be assessed for a broader class of glass-forming liquids, including polymers and molecular liquids. A good starting point for this simulation campaign could be the molecular liquid studied by Shiraishi et al.¹⁰, who found that the separation between fast and slow processes was due to a two-barrier structure of the potential energy landscape. Hopefully, this will also trigger a shift to computational studies of molecular models of experimental relevance.

Daniele Coslovich  

Dipartimento di Fisica, Università di Trieste, Trieste, Italy.

 e-mail: dcoslovich@units.it

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