Small-beam diffraction measurements for understanding local structure and local dynamics in glasses

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In centrosymmetric crystals, each atom is at a centre of symmetry and thus at mechanical equilibrium, with the forces balanced by all its neighbours. Under an applied force, the particles undergo affine displacements that are proportional to this force until the elastic limit is reached, and topological defects are created. In dense glasses with isotropic interparticle interactions (e.g. metallic and colloidal glasses) the local environment around any given particle is generally not a symmetric polyhedron and the imbalance of forces results in irreversible, non-affine displacements [1]. The nature of the local structures that undergo these displacements under mechanical loading, or “flow defects”, in glasses is not known.

In this study, we make a direct link between local stability and local structure in a glass using scanning microbeam small-angle x-ray scattering (Figure 1 A) [2]. We employ micro-x-ray cross-correlation measurements [3] to resolve spatial variations in local structural dynamics (Figure 1 B at time delays of Δt = t and Δt = 2t). We analyze the angular symmetries and distribution of intensity in the speckle diffraction patterns to measure the local degree of centrosymmetry and the local structural anisotropy or strain (Figure 1B). Our measured maps of softness and structure demonstrate that even though local stability and local centrosymmetry fluctuate at the length scale of a single polyhedron, the structural centrosymmetry is still a strong predictor for mechanical stability in glasses. Furthermore, we examine local stability and structure during glass aging and deformation and find that coordinated local structural transformations to lower symmetry structures are central to these phenomena [2]. This provides new particle-level insight into aging and deformation-induced localization of strain in “shear bands”, the precursor to brittle mechanical failure in glasses. Similar approaches using scanning electron nanodiffraction may give insights into atomic glasses [4].

Figure 1. A Microbeam small-angle x-ray scattering of a colloidal glass produces rich speckle diffraction patterns that reflect the local structure. A single polyhedron has a diameter of 1 µm. B Despite fluctuations, maps of local stability and the local degree of centrosymmetry and local anisotropy (here normal strain or dilatation/contraction) show strong cross-correlations.


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