Nanoscopic Dynamics in a Supercooled Liquid from Electron Correlation Microscopy

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The dynamics of atomic rearrangements in supercooled liquids control behavior including nucleation of crystalline phases and the glass transition. There is significant but indirect evidence that these dynamics are spatially heterogeneous at the nanometer scale. That is, different nanoscopic volumes of the liquid exhibit significant different structural relaxation times t, where τ is the time it takes for atoms in the liquid to significantly rearrange, for example by gaining or losing a neighboring atom. At the macroscopic level, τ is connected to the liquid viscosity and to diffusion.

We have developed an electron microscopy technique based on time-resolved electron nanodiffraction called electron correlation microscopy (ECM) to measure relaxation times in liquids *in situ* in the STEM with sub-nanometer spatial resolution [1]. ECM experiments on $Pt_{57.5}Cu_{14.7}Ni_{5.3}P_{22.5}$ bulk metallic glass nanowires [2], heated above the glass transition temperature T_g inside the microscope show direct evidence of spatially heterogeneous dynamics. The size of the domains increases and their relaxation time slows as the sample cools from near the crystallization temperature toward T_g, in agreement with microscopic theories of the glass transition. We also observe a ~1 nm thick surface layer with dynamics an order of magnitude faster than the bulk. Fast near-surface dynamics provides an explanation for the surface crystallization of the nanowires which does not require heterogeneous nucleation sites.

New ECM experiments with a fast direct electron detection camera have the potential to rigorously test microscopic theories of the glass transition and provide new insight into relaxation dynamics in the solid glassy state. Challenges and opportunities for direct electron ECM experiments will be discussed.

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