Fragile-to-strong crossover and non-monotonic decoupling of diffusion from structural relaxation in a binary metallic glass-forming alloy

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Abstract:

The glass-forming ability of liquids is now commonly characterized in terms of 'fragility'[1] which is defined with the help of T_g/T dependence of the viscosity (η) or structural relaxation time (τ). It is classified as "strong" if η or τ follows an Arrhenius law and "fragile" when it deviates from it, showing an upward curvature or super-Arrhenius behavior. While most of the glass-forming liquids fall in the ambit of this scheme, the concept of fragility remains limited as it implies that the fragility correctly describes the super-Arrhenian behavior over the entire temperature range of interest, say from the melting temperature T_m to the glass transition temperature T_g where τ is supposed to diverge.

Contrary to this prevailing view, it was conjectured from the analysis of the temperature dependence of η for a variety of liquids that they display a fragile-to-strong crossover (FSC) in the undercooled region. Such crossover can be linked to changes in dynamic regimes and relaxation processes[2,3]. A recent extended mode-coupling theory (MCT) study predicts a scenario for the FSC in which τ and the self-diffusion constant(D) cross over from a super-Arrhenius to an Arrhenius behavior.[4] It also provides a possible explanation of the FSC observed in systems where the existence of Widom line (like the case of water) is unlikely.

Our recent investigations of a Cu-Zr bulk metallic glass forming alloy[5] reveal that: (i) the alloy exhibits a non-monotonic decoupling of *D* and τ as observed in case of supercooled water despite the difference in the intermolecular interactions compared to this system, (ii) the alloy exhibits a crossover from Stokes-Einstein (($D \sim (\tau/T)^{-1}$) to fractional Stokes-Einstein ($D \propto (\tau/T)^{-\zeta}$) with exponent $\zeta \approx 0.6$. A weak first-order transition, associated to the FSC, has also been observed in the undercooled region.

The talk is intended to discuss these ideas.

References:

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