Structural transitions in metallic and semiconducting glassformers: some phenomenological relationships with connections to phase change switching materials

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Abstract:

One of the more surprising developments in the area of metallic glassformers in recent years has been the suggestion by Zhang et al (J.Chem. Phys. 133, 014508 (2010) that a large number of metallic glassformers exhibit some sort of structural transition, manifested as a change from fragile liquid character at high temperature to strong liquid character at lower temperature approaching the glass transition. This occurs in the temperature range where the crystallization is particularly rapid, and the two are no doubt closely related. This sort of behavior was previously considered a characteristic only of certain highly anomalous "tetrahedral liquids" like water, and liquid silicon. In the latter, the transition is also a metal-to-semiconductor transition as suggested long ago by Turnbull and coworkers for Si and Ge, before confirmation by the simulations of Grabow, Sastry and their coworkers.

In the metallic glassformer case, of course, the transition cannot be assigned to any electron localization phenomenon, while in the water case, there is no hint of delocalized electrons, so we must search for some more general explanation. We note that in the cited cases, the glass transition occurs below the liquid-liquid phase transition, where the liquid exhibits "strong" liquid characteristics. Analogous behavior is seen in the case of the strong extreme of rotator phase crystals, where the high temperature limit is a lambda transition of theoretically well-understood character. In liquids, lacking crystalline order, the lambda form is only observed at a particular pressure, where it is a critical point terminating a line of first order liquid-liquid transitions. At different pressures the density fluctuations may be damped out before becoming critical, or may be preempted by a first order transition. We illustrate these features using "potential tuning" MD of the Stillinger-Weber model for liquid silicon. Finally, we link these ideas to known density anomalies (with supercritical appearance) in supercooled liquid Te, and stable liquid As2Te3, and discuss how they might find practical manifestation in the fast switching phenomena of "phase change" materials (which contain these two elements and Ge in similar proportions). (Angell, in Physics and Applications of Disordered Materials (Honoring Stan Ovshinsky on his 80th: editor M. Popescu) pp 1-18 (2002)).