Liquid and Glass Structures and Their Influence on Phase Transitions

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Recent developments in novel levitation methods have led to a renewed interest in the structures of equilibrium and supercooled liquids. An identification of local and medium-range ordering and the influence of this ordering on thermophysical properties and liquid phase transitions are key questions of fundamental interest. Structural studies of phase transitions from equilibrium and metastable liquids are also of practical note, allowing regions of the alloy phase diagram to be quickly and accurately determined.

Employing the technique of electrostatic levitation [1], we have measured metallic and semiconductor liquid x-ray diffraction patterns as a function of supercooling. The liquid order was characterized in terms of a dominant local cluster model and by Reverse Monte Carlo (RMC) fits to the diffraction data. The topologies of the RMC structures are expressed in terms of their bond orientational order (BOO) parameters and Honeycutt-Andersen (HA) indices. These studies have led to new insights into the structures of liquids and the phase transitions that occur within them. For example, our studies of liquid Si show no evidence for an increase in the A4 (diamond cubic) order from the A5 (white tin) order that is characteristic of the high temperature liquid, raising doubts about the existence of a predicted liquid/liquid phase transition in the supercooled liquid. In supercooled transition metal alloy liquids, many develop significant icosahedral short-range order (ISRO) [2]. This is true even in elemental transition metal liquids, although there it can be significantly distorted [3,4]. The HA and BOO analysis of the RMC structures, however, demonstrate that while ISRO is dominant, a significant number of the ordered atoms also sit in other environments, including distorted ISRO and BCC and FCC-like structures [5].

The developing ISRO couples to the first-order phase transition, lowering the nucleation barrier for the icosahedral quasicrystal in these transition metal alloys, with the ordered regions acting as a template for the formation of the new phase. Measurements of the S(q) for a $Zr_{59}Ti_3Cu_{20}Ni_8Al_{10}$ liquid and glass, coupled with the first quantitative measurements of the time-dependent nucleation rate in a metallic glass (in this case for the nucleation of an icosahedral quasicrystal), demonstrate that a developing icosahedral short-range order accompanies the glass transition. These and related points will be discussed.

References

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