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Coupled Nucleation Processes in Metallic Liquids and Glasses[1]

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Nucleation processes in condensed systems are often more complicated than expected from classical theory considerations. For example, our recent studies of glasses and deeply supercooled liquids demonstrate that the short- and medium-range order play an important role in the nucleation pathway. High-energy X-ray diffraction data from electrostatically levitated transition metal and alloy liquids demonstrate the frequent development of icosahedral short-range order (ISRO) with supercooling. This ordering has significant consequences for crystallization and vitrification of the liquids. It makes it difficult to nucleate ordered crystal phases, confirming a half-century old hypothesis by Frank. Measurements of the density and surface tension in several supercooled liquids suggest that it may be associated with a liquid/liquid phase transition. Quantitative measurements of the time-dependent nucleation rate in a $Zr_{59}Ti_3Cu_{20}Ni_8Al_{10}$ metallic glass and associated structural studies of the supercooled liquid demonstrate that it increases through the glass transition, providing support for a frustration model of the glass transition. In a Ti-Zr-Ni liquid the ISRO lowers the barrier for a metastable icosahedral quasicrystal, blurring the distinction between homogenous and heterogeneous nucleation. Our studies and those of others suggest that the nucleation of the ordered phase can be coupled with liquid phase transitions, including high order transitions. Coupling between other processes is also common for nucleation. For example, the coupling between the stochastic fluxes of interfacial attachment and long-range diffusion in the nucleation step can be critical when the initial and final phases have different chemical compositions. The implications of coupled nucleation processes on phase formation, stability and nanoscale crystallization are discussed.

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