Heterophase liquid states: thermodynamics, structure, dynamics

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Abstract:

Heterophase fluctuation is a transient solid-like or fluid-like cluster (fluctuon) characterized by a short-range order (SRO). Non-crystalline solid-like fluctuons are not compatible with the translational-invariant tiling of the space. For this reason joining of the solid-like fluctuons is accompanying by deformations or formation of topological defects. This property is known as structural frustration [1]. Increase of the free energy due to frustration is the frustration parameter, g.

Thermodynamics, structure and dynamics of the heterophase liquid states is described within the framework of the model of heterophase fluctuations taking into account multiplicity of SRO of solid-like fluctuons with multicomponent order parameter [2-4] . The frustration parameter is introduced [4] in a different manner than that in [1]. In the two-component case the model is isomorphic to many known two-state models. In general case it is isomorphic to Ising model with multicomponent spin and “external field” expressed in terms of the thermodynamic parameters of fluctuons. Equations of the state describe temperature dependence of the fluctuon fractions. The temperature range of heterophase liquid state is scaled by g, its lower and upper boundaries separate homophase glassy and fluid states. Parametric phase diagram on the plane (T,g) contains the critical point on the phase coexistence line at low g . Parametric evolution curves show thermal history of heterophase state. Glass residual entropy consists of mixing and frustrational entropies.

Structure factor of the “mosaic” states with and without Fischer cluster in terms of the order parameter is obtained. Relations of α-relaxation, fragility, ultraslow mode spectrum and Fischer cluster equilibration time with the order parameter and thermodynamic coefficients are described. Coincidence of the temperature dependences of the two-component order parameter obtained independently from analysis of experimentally measured enthalpy, structure factor and α-relaxation time of salol demonstrates adequacy of the theory.

References:

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