Hidden structural ordering in a supercooled liquid: Intrinsic link between glass transition and crystallization

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Recently it has been revealed that when approaching the glass transition temperature Tg, liquid dynamics not only drastically slow, but also become progressively more heterogeneous. From our simulations and experiments of six different glass-forming liquids[1-5], we find that the heterogeneous dynamics is a result of critical-like fluctuations of static structural order, contrary to a common belief that it is purely of dynamic origin: The 'static' correlation length and susceptibility of a structural order parameter exhibit Ising-like power-law divergence towards the ideal glass-transition point[5]. However, this structural ordering accompanies little density change over a long range, which explains why it has not been detected by the static structure factor so far. Our results suggest a far more direct link than thought before, between glass transition and critical phenomena: Glass transition may be a novel type of critical phenomena, where a structural order parameter is directly linked to slowness.

This critical-like structural ordering also has an important implication on crystallization, which intrinsically takes place in a supercooled liquid state. For hard-sphere-like colloidal liquids, for example, we reveal that in a supercooled liquid state, medium-range crystalline bond orientational order of the hexagonal close pack structure grows in its size and lifetime with increasing the packing fraction[5,6]. We reveal that nucleation of crystals preferentially occurs in regions of high medium-range order, reflecting the low crystal-liquid interfacial energy there[6]. These findings may shed new light not only on the fundamental nature of glass transition, but also the mechanism of crystal nucleation. The relation of these findings to our two-order-parameter model of glass transition[7,8] will also be discussed.

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