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General nature of liquid-liquid transition in aqueous organic solutions

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Contrary to the conventional wisdom that there exists only one unique liquid state for any material, there are growing experimental and numerical pieces of evidence for the existence of more than two liquid states in a single component substance. The transition between them is called liquid-liquid transition (LLT). LLT has attracted considerable attention because of its importance in the fundamental understanding of the liquid state. However, the physical nature of the transition has remained elusive. Particularly for water, the possible existence of LLT has special implications not only on its fundamental understanding, but also on a link of various thermodynamic and transport anomalies with critical anomaly associated with LLT. Here we reveal that 14 aqueous solutions of sugar and polyol molecules, which have an ability to form hydrogen bonding with water molecules, exhibit liquid-liquid transitions. We find evidence that both melting of ices and liquid-liquid transitions in all these aqueous solutions are controlled solely by water activity, which is related to the difference in the chemical potential between an aqueous solution and pure water at the same temperature and pressure. Our theory shows that water activity is determined by the degree of local tetrahedral ordering, indicating that both phenomena are driven by structural ordering towards ice-like local structures. This has a significant implication on our understanding of the low-temperature behaviour of water.

Keywords: liquid-liquid transition, water and aqueous solution, supercooled liquids and glasses