# Relationship between the glass transition and medium-range crystalline order

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Some liquids do not crystallize below the melting point and enter into a supercooled liquid state upon cooling. During this process, the structural relaxation time drastically increases with a decrease in the temperature and finally they become a glassy state. It has not been clarified what physical factors control the glass transition phenomena despite intensive efforts over more than several decades. All existing theories presuppose the avoidance of crystallization because they consider that crystallization is nothing to do with the slow dynamics of a supercooled liquid. So these theories cannot answer the fundamental question of what controls the ease of vitrification.

Differently from these physical views, we thought vitrification is intrinsically linked to crystallization and proposed "two order parameter model"[1]. According to this model, vitrification and the resulting slow dynamics are due to the frustration between long-range density ordering toward crystallization and short-range bond ordering toward the formation of locally favored structures.

Based on this idea, we introduced a new type of interaction potential that can directly control the tendency of short-range bond ordering, namely, the strength of frustration against crystallization. Thus we succeeded in developing a model that can cover crystallization to vitrification just by changing the degree of frustration and found that there is a clear correlation between the slow dynamics and the frustration (see Figure 1).

We also found that there exist medium-range crystalline ordered areas (MRCO) in supercooled liquids which are highly correlated with dynamic heterogeneity (see Figure 2).

Our simulation results indicate that the degree of MRCO controls the slow dynamics of supercooled liquids and there is an intrinsic link between glass transition and crystallization.

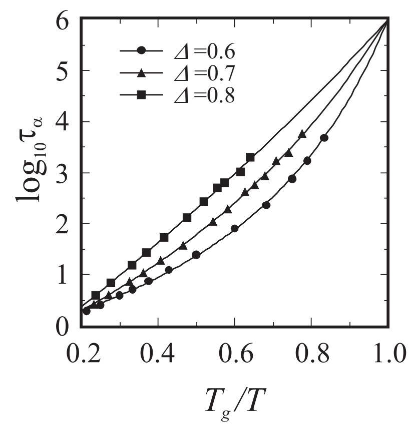


Figure 1: Angell plot for several liquids with different degrees of frustration. $\Delta$ represents the strength of frustration against crystallization. It is clear that the stronger the frustration is, the stronger the liquid is. This result is consistent with the prediction of two order parameter model.

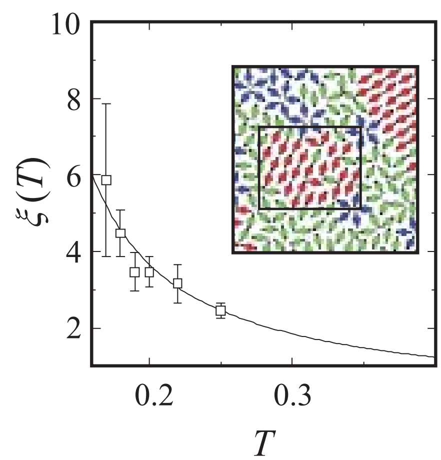


Figure 2: Temperature dependence of $\xi$ (the size of MRCO). This is fitted by $(T - T_0)^{-1}$ ($T_0$ is obtained by the fitting of the Vogel-Fulcher relation to T-dependence of $\tau_\alpha$). Inset: The snapshot of a supercooled liquid ($T = 0.17$). The arrow of particle is the directional vector of the particle. The area in open square is the medium range antiferro crystalline ordered area (MRCO). Note that this structure has long lifetime compared to the relaxation time of liquids but is not static structure.

## References

[1] H. Tanaka, J. Chem. Phys. **111**, 3163 (1999).