Title: Relationship between dynamic heterogeneity and medium-range crystalline order

Author(s): Kawasaki, Takeshi; Araki, Takeaki; Tanaka, Hajime

Citation: 物性研究 (2006), 87(1): 152-153

Issue Date: 2006-10-20

URL: http://hdl.handle.net/2433/110601

Type: Departmental Bulletin Paper

Textversion: publisher

Kyoto University
Relationship between dynamic heterogeneity and medium-range crystalline order

Institute of Industrial Science, University of Tokyo
Takeshi Kawasaki, Takeaki Araki and Hajime Tanaka

Upon cooling, some liquids do not crystallize below the melting point, but instead become a glass at the glass transition temperature $T_g$. The most remarkable characteristic of the glass transition is that the dynamics of the liquids becomes significantly slower toward $T_g$ with keeping its amorphous structure. Although many theoretical and numerical studies have been devoted to the origin of the glass transition, it remains unclear. Recently some study reported the existence of the dynamic heterogeneity in a glassy state and claimed that it might play a very important role in liquid-glass transition [1, 2]. Further some experimental and numerical studies reported that the cluster of the medium-range crystalline order does exist in a supercooled liquid state [3, 4]. Here we study the relationship between the dynamic heterogeneity and medium-range crystalline order by means of 2D Brownian dynamics simulation of polydisperse colloidal dispersion.

Polydisperse colloidal dispersion have attracted considerable interests as a model glass-forming liquid [5]. For a monodisperse system, a system just crystallizes from a liquid state as the volume fraction of colloidal particle, $\phi$, increases. On the other hand, a system of a high polydispersity does not crystallize even for large $\phi$, and instead its dynamics slows down with the increasing $\phi$. In other words, it enters a disordered glassy state. Since the polydispersity is regarded as the frustration upon the crystallization of liquids, we can control the glass-forming ability of a colloidal system by changing the degree of polydispersity. In our simulations, colloidal particles interact repulsively with each other as

$$U(r_{ij}) = \begin{cases} 4\varepsilon \left\{ \left( \frac{\sigma_{ij}}{r_{ij}} \right)^{12} - \left( \frac{\sigma_{ij}}{r_{ij}} \right)^{6} + \frac{1}{4} \right\} & \text{(for } 0 < r_{ij} < 2^{\frac{1}{6}} \sigma_{ij}) \\ 0 & \text{(otherwise)} \end{cases}$$

1E-mail: kawasaki@iis.u-tokyo.ac.jp
where $r_{ij} = |\vec{r}_i - \vec{r}_j|$, $\vec{r}_i$ is the position of particle $i$. $\sigma_{ij} = (R_i + R_j)/2$, where $R_i$ is the diameter of particle $i$ given by Gaussian distribution whose polydispersity is defined as $\Delta\sigma = \sqrt{\sum_i (R_i - \bar{R})^2/(N_p - 1)}$, where $\bar{R}$ is the average of the particle radii and $N_p$ is the particle number.

Our simulations revealed the existence of the medium-range crystalline order in supercooled liquid states of the polydisperse colloidal dispersions by calculating the six-fold positional order $\Psi_6$. The six-fold positional order $\Psi_6$ is given as

$$
(\Psi_6)_i = \frac{2}{n(n-1)} \sum_{k=1}^{n} \sum_{l=k+1}^{n} e^{i6\theta_{kl}},
$$

(2)

where $j = \sqrt{-1}$, $n$ is the number of nearest neighbors of particle $i$, and $\theta_{kl}$ is angle formed by the junction of $\vec{r}_k - \vec{r}_i$ and $\vec{r}_l - \vec{r}_i$ where particle $k$ and $l$ are nearest neighbors of particle $i$. And we found that there is a strong correlation between the six-fold positional order and the motion of each particle. The particles are trapped more and less in clusters of medium-range crystalline order. While, those out of the cluster can move smoothly. The clusters have typically 10 times larger life time than the relaxation time of the liquid $\tau_\alpha$. Therefore it is considered that this correlation is the origin of the dynamic heterogeneity.

References


