Free energy landscape of glass-forming substance

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Free energy landscape picture is believed to provide a unified understanding of the thermodynamic and dynamic singularities near the glass transition temperature. However, the landscape has not explicitly been defined nor constructed on the basis of the microscopic Hamiltonian. Because of this lack of explicit theory, the dynamics in the free energy landscape has not been analyzed so far.

Exploiting the density functional theory, we give a clear definition of the free energy landscape as a function of a particle configuration. By using the 3-D randam configuration of hard spheres, we obtain the free energy landscape for a relaxation process characterized by a string motion. Figure 1 shows the free energy landscape as a function of the displacement of the specific particle. It turns out that the system transits the adjacent basin via the saddle point.

We also determine the size of the cooperatively rearranging region (CRR), N_{CRR} from the free energy landscape. The CRR corresponds to the particles which are moved at the saddle point from the configuration at the minimum of the original basin.

N_{CRR} is measured in the following way: (i) prepare the spherical shell made by fixing the particles (Figure 2); (ii) calculate the free energy landscape when we force to move 1 particle (Figure 2); (iii) if the system transits the adjacent basin, N_{CRR} is smaller than those in the spherical shell, and otherwise, N_{CRR} is larger than those in the spherical shell; (iv) by changing the inner diameter of the spherical shell, one can estimate N_{CRR}.

Figure 3 shows the density dependence of the N_{CRR}. It turns out that N_{CRR} is increased as the density is raised, and that the density dependence of N_{CRR} can be fitted by the Vogel-Fulcher law.

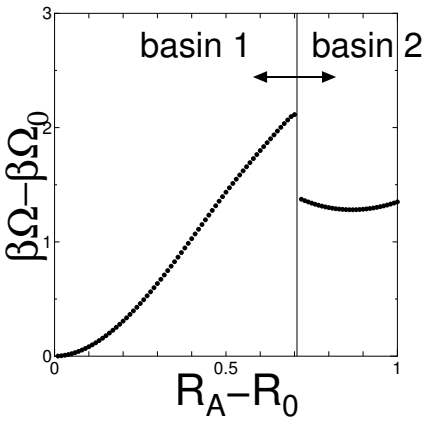

Figure 1: Free energy landscape as a function of the displacement of the specific particle.

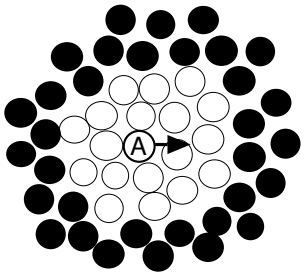

Figure 2: Model to calculate N_{CRR}.

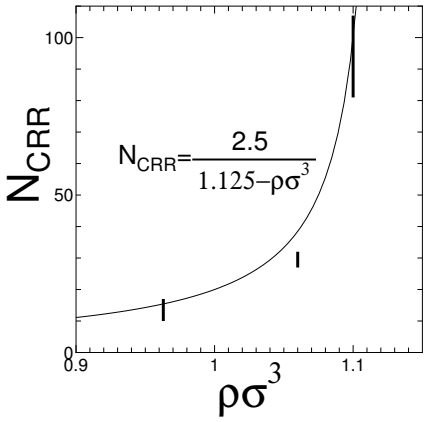

Figure 3: density dependence of N_{CRR}.