Electronic Supplementary Information

I. SELF AND DISTINCT DIFFUSION COEFFICIENTS

We have analyzed the simulation data for the intermediate scattering function $f_{s/d}(q, t)$ for wavenumbers $q\sigma$ between 2 and 10 according to the fitting ansatz:

$$f_{s/d}(q, t) = \sum_{i=1}^{2} A_{i}^{s/d}(q) \exp(-q^{2}D_{i}^{s/d}(q)t) (1)$$

In general, the results for the self part $f_{s}(q, t)$ showed moderate statistical fluctuations and could be fitted reliably with the formula above. The statistical fluctuations in the distinct part $f_{d}(q, t)$ (for fixed $t$) differed a lot and in an unsystematic manner between different $q$. In the results shown below we included those $q$–points for which fits delivered a maximum error of 50% for the diffusion coefficients.

Fig. 1 shows the results for $D_{i}^{s/d}(q)$ for state points along path I (packing fraction $\Phi = 0.1$, bond concentration $c_{b} = 2$ and varying temperature $T = 0.14...0.10$). The main conclusion from the plots is that there is a robust separation between a “fast” and a “slow” process, with the corresponding diffusion coefficients ($D_{2}$ and $D_{1}$) differing by a factor of about 10. The variation of the static structure factor for $q\sigma = 2...10$ (see Fig. 5 in the main paper) is rather weak, therefore it is justified to compute an average diffusion coefficient (see Fig. 8 in the main paper) by the error weighted mean value of the fitted diffusion coefficients $D_{1}(q)$ and $D_{2}(q)$, respectively:

$$\bar{D}_{i}^{s/d} = \frac{\sum_{q} D_{i}^{s/d}(q)/\epsilon(q)^{2}}{\sum_{q} 1/\epsilon(q)^{2}} ,$$

where $\epsilon(q)$ is the relative error in $D_{i}^{s/d}(q)$ as obtained by the fit. Note, that we did not take into account any $q$–dependence of the diffusion coefficients which might be suggested by the figures for the self part (Fig. 1, right column). The systematic rise at low $q$ is due to the limited simulation time, such that the decay of the intermediate scattering function is not fully present in the data. Likewise for larger $q$ the time bin widths are too long to fully resolve the fast decay, also resulting in a systematic rise at larger $q$. This, in turn, results in the systematic overestimation of the self diffusion coefficients at low $q$. To fully resolve the low $q$ coefficients, much longer simulation runs would be needed, being beyond the scope of
this work. Ignoring the systematic rise at low $q$, a constant self diffusion coefficient is well justified by the data.

Fig. 2 shows the results for $D_{s/d}(q)$ for state points along path II (packing fraction $\Phi = 0.05$, temperature $T = 0.1$ and varying bond concentrations $c_b = \{0, 1.2, 2\}$). For $c_b = 0$, the system is purely repulsive, therefore the average diffusion coefficients $D_1$ and $D_2$ are rather close and do not support the existence of two time scales. For nonzero bond concentration, the same robust separation between a “fast” and a “slow” process is found as for path I.

Fig. 3 shows the ratio of “fast” and “slow” distinct diffusion coefficients along the two paths as a function of $q$. There is a considerable scatter of this ratio but a majority of the data points are in the range 10...30.
FIG. 1. Fitted diffusion coefficients along path I (packing fraction $\Phi = 0.1$, bond concentration $c_b = 2$ and varying temperature $T$) from the intermediate scattering function: Distinct part (left column), self part (right column). Horizontal lines are mean values of the respective data points.
FIG. 2. Fitted diffusion coefficients along path II (packing fraction $\Phi = 0.05$, temperature $T = 0.1$ and varying bond concentration $c_b$) from the intermediate scattering function: Distinct part (left column), self part (right column). Horizontal lines are mean values of the respective data points. Note that the scatter in the upper right figure results from attempting to fit a double-exponential to data which contain essentially a single exponential decay (as can be seen from the proximity of the averages).
FIG. 3. Ratio $D_2/D_1$ of the diffusion coefficients (distinct part) belonging to the fast and the slow process as a function of $q$. Left panel: path I. Right panel: path II.