Electronic Supplementary Material (ESI) for Soft Matter.

Dynamic properties of different liquid states in systems with competing interactions studied with lysozyme solutions

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Supplementary Information

SANS data of lysozyme samples at different concentrations and different temperatures

The absolute scattering intensities, $I(q)$, are normalized by the protein volume fraction, ϕ, after subtracting the background scattering intensity, B, which is quantified by fitting the absolute scattering intensities in Figure S0a in the region between q-values of 0.4 to 0.55 Å^{-1}. Raw SANS data is provided in the ESI. To be consistent with previous studies,[1–3] the volume fraction of these samples are estimated using the skeleton density unless otherwise stated explicitly.

It is well known that decreasing the temperature increases the attraction strength between proteins so that the inter-protein structure factor, $S(Q)$, is strongly temperature dependent, while the form factor, $P(Q)$, remains largely unaffected by changing solution conditions. This is confirmed by the results shown in Figure 1. The normalized intensities for all concentrations and temperatures overlap at relatively high q-values, which probe the length scale of individual proteins, indicating that the globular structure of lysozyme is maintained under all solution conditions. Variations in the q-dependence of normalized $I(q)$ at low q-values is therefore related to changes in inter-protein interactions.

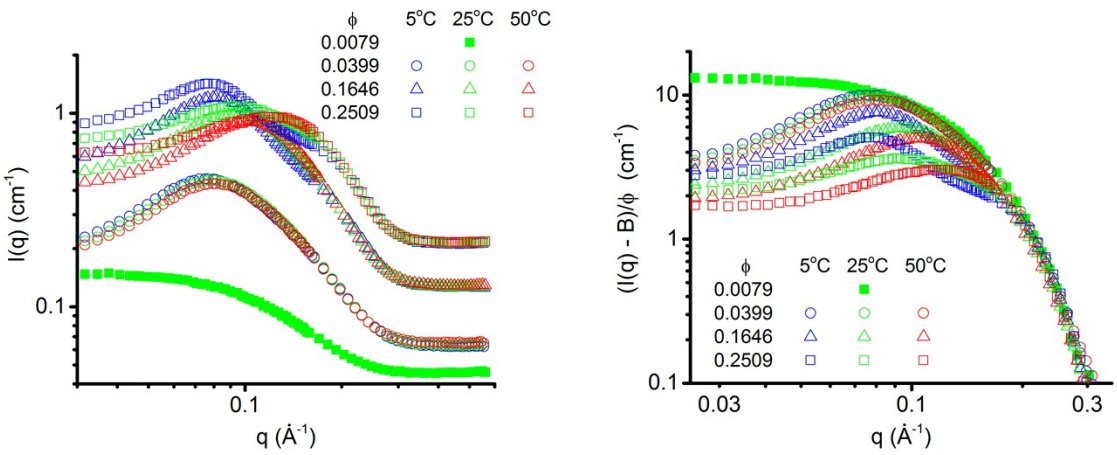

Figure S0. (a) The absolute intensity from SANS experiments is plotted as a function of q-value for the same lysozyme sample conditions as shown in Fig.1 of the main text. **(b)** Absolute scattering intensities obtained from SANS experiments scaled by their corresponding background and volume fraction are plotted for three lysozyme concentrations at three temperatures each (open symbols) along with a low concentration sample representative of the form factor (closed symbols).

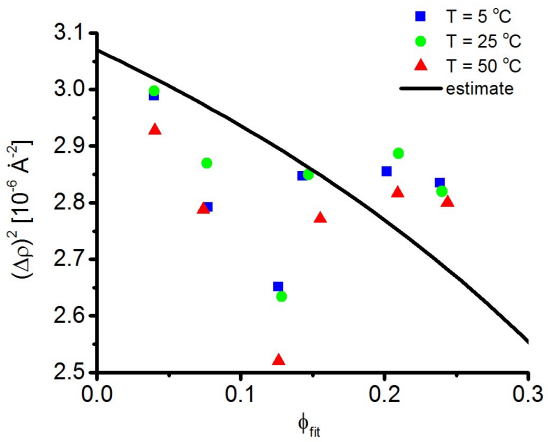

Figure S1. The neutron SLD contrast of lysozyme shifts with protein concentration due to hydrogen-deuterium exchange when suspended in heavy water. This shift is explicitly accounted for when fitting the SANS data, which utilized the values shown here (symbols) compared with the shift estimated by assuming 7% of hydrogens in lysozyme are labile and free to exchange (line).

Monte Carlo Simulations to verifying the IET theory and the percolation transition

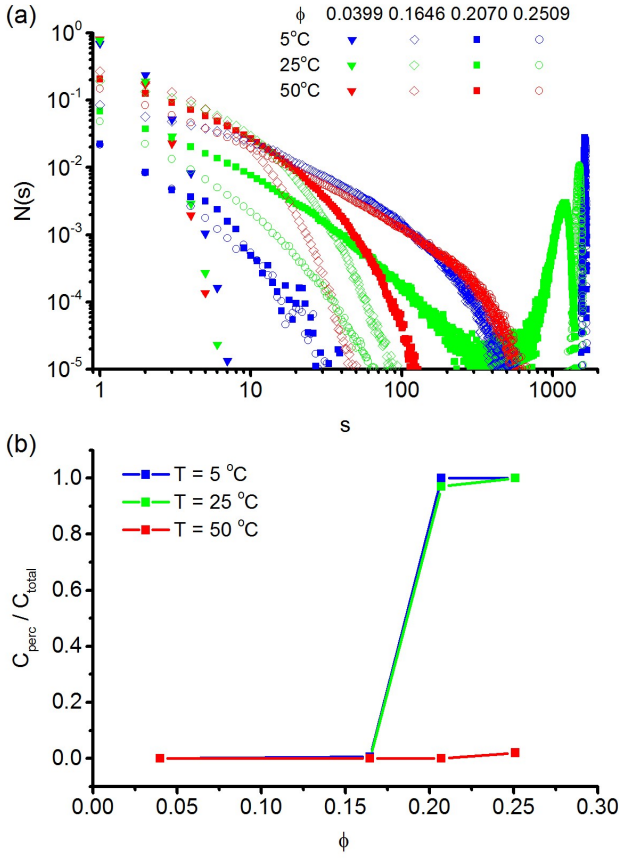

Figure S2. (a) The cluster size distributions are shown for each of the state points visualized using MC simulations, demonstrating the onset of large, system-spanning cluster formation at high volume fractions and low temperature. Some of the data is replotted from our previous pulication.[4] (b) The fraction of states that have a percolated cluster is plotted as a function of volume fraction, from which the percolation transition is determined to be when C_{perc}/C_{total} is above 0.5, as defined previously.

Estimation of the characteristic diffusion time of lysozyme at different temperatures

The characteristic diffusion time depends on the temperature and solution viscosity. Table S1 show the calculated characteristic diffusion time defined in the main text.

Table S1. Summary of characteristic times of diffusion, t_D, for lysozyme at each temperature studied.

T (°C)	t_D (ns)
5	35.6
25	18.2
50	10.0

Summary of gel and glass transition estimates

The gel and glass transitions corresponding to lysozyme samples at temperatures of 5 ^{o}C, 25 ^{o}C, and 50 ^{o}C, which are quantified by fitting the volume fraction dependence of microrheology and NSE data with a power law function, are summarized below:

T (^{o}C)	τ_B	ϕ_g	σ	ϕ_{gel}	σ
5	0.183	0.487	0.020	0.416	0.045
25	0.457	0.519	0.022	0.416	0.050
50	1.209	0.578	0.029	0.467	0.075

Estimation of the characteristic

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2. F. Cardinaux, E. Zaccarelli, A. Stradner, S. Bucciarelli, B. Farago, S. U. Egelhaaf, F. Sciortino, and P. Schurtenberger, *J. Phys. Chem. B*, 2011, **115**, 7227–7237.

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