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Mesoscopic Aggregation in Protein Solutions[1]

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Long-lived mesoscopic clusters of a dense protein liquid are observed in concentrated solutions of numerous proteins. These clusters are a necessary kinetic intermediate for the formation of solid aggregates of native and misfolded protein molecules. We propose a novel physicochemical mechanism, by which the clusters consist of an off-equilibrium mixture of single protein molecules and long-lived protein-containing complexes. The puzzling mesoscopic size of the clusters is determined by the lifetime of the complexes and their diffusivity. We have predicted and observed a number of interesting kinetic and thermodynamic behaviors that are associated with the mesoscopic clusters. These behaviors include: (a) Ostwald-like ripening of the clusters (b) a crossover to long-range density fluctuations at high concentrations; (c) a universal, diffusion-like scaling of the autocorrelation function of light scattered off the protein solution; (d) non-trivial dependencies of the cluster size and volume fraction on the protein concentration in the solution. Our analysis of the anisotropic Coulomb interactions suggests for mesoscopic clusters to form in lysozyme solutions, protein molecules must undergo conformational changes.

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