Multiple Pathways in NaCl Homogeneous Crystal Nucleation

Supporting Information

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Additional Figures

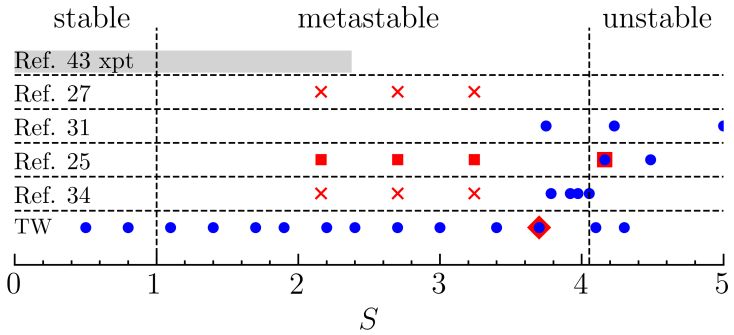

Figure S1: Supersaturation (S) levels adopted in recent simulations (using the force field from Ref. 36 at room temperature) and experiments, as indicated by the references to works in the main paper. 'TW' refers to the supersaturations simulated in this work. Circles, crosses and squares indicate brute force MD, seeded MD and forward flux sampling simulations, respectively. The diamond indicates the combination of seeded MD and metadynamics adopted in this work to study nucleation at $S = 3.7$. The grey bar indicates the range of S measured in experiments of levitated droplets of NaCl(aq) solutions where liquid-like clusters were detected at the high end of S.

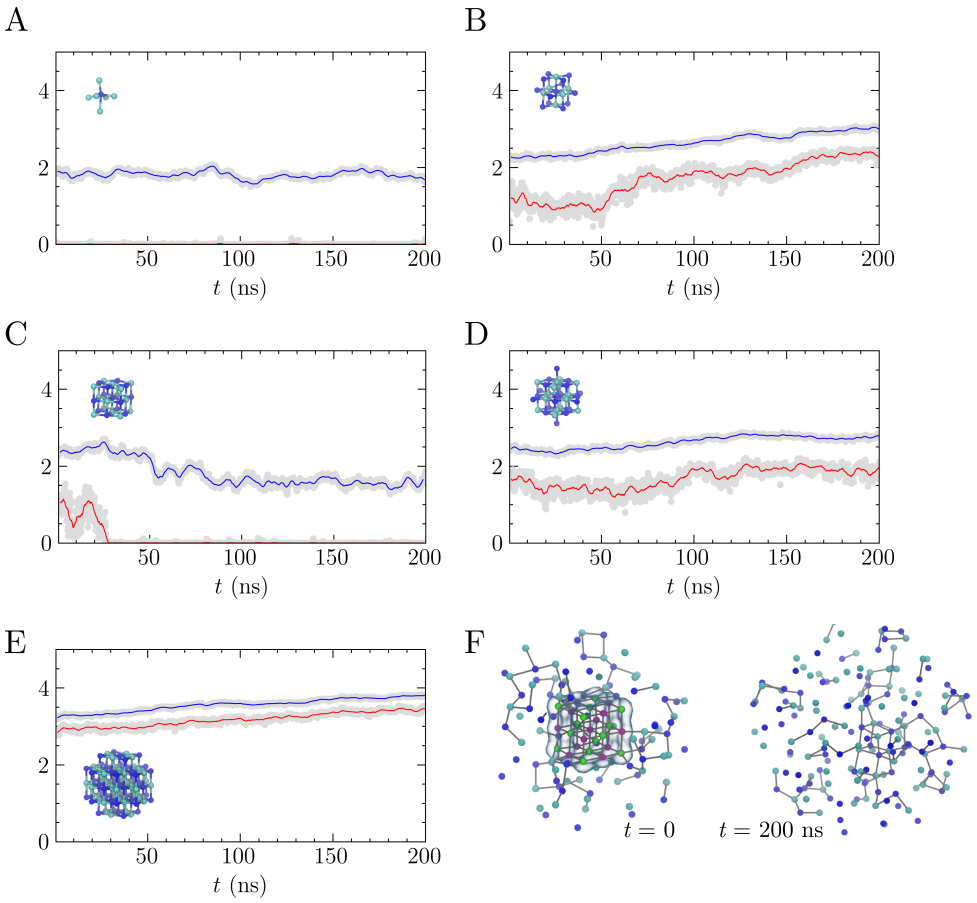

Figure S2: Simulations of crystalline seeds in solution where the diameter of the seeds was $0.6 - 1.4$ nm in 0.2 nm increments for A–E. The time series for n^{sph} (blue) and $n^{sph}(q6)$ are provided. Inset are the initial configurations for the seeds which were immersed into solution. F shows example configurations for the species in solution from the simulation in C at $t = 0$ and 200 ns. Blue and cyan spheres represent Na^+ and Cl^-, respectively, and the first sphere coordination between ions is highlighted by the grey lines. The ions in the initial crystalline seed in F are highlighted by the purple (Na^+) and green (Cl^-) spheres.

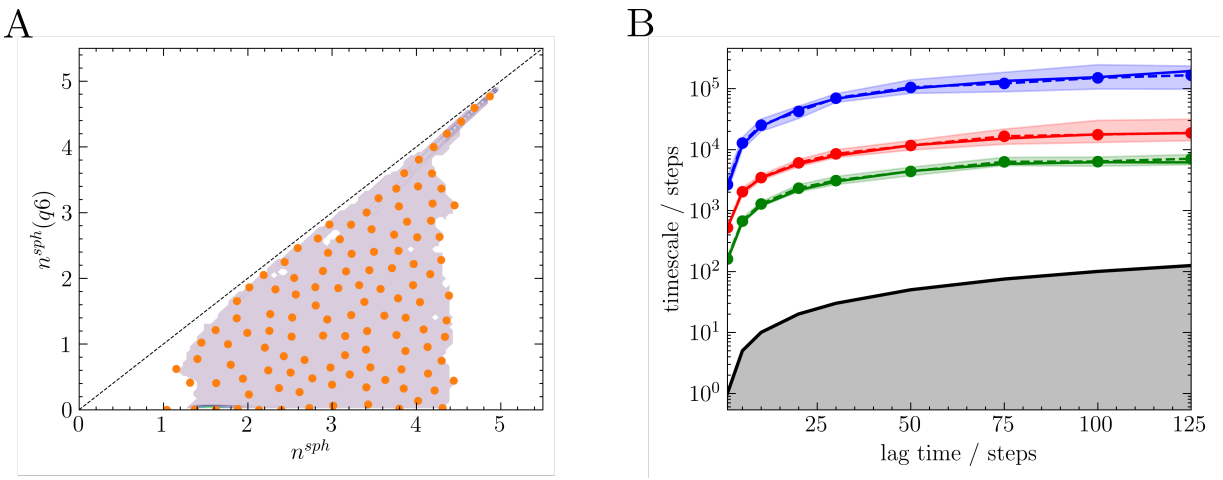

Figure S3: A: Points show the partitioning of the reaction coordinate into ~ 100 states used in the Markov State Model described in the main text. B provides the timescale for different lag times between the states.

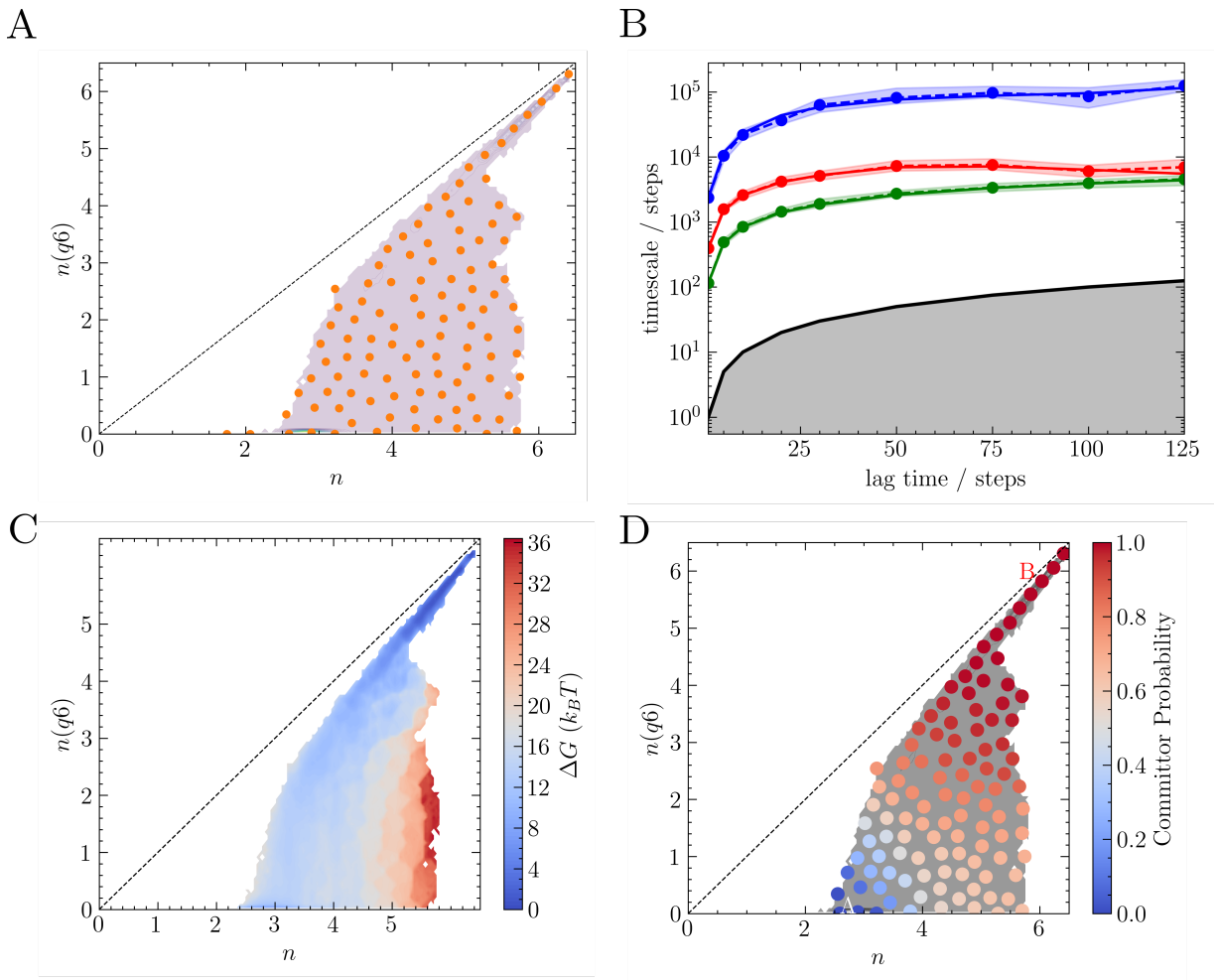

Figure S4: State points (A), lag times (B), free energies (C) and committor probabilities (D) from the MSM characterising transitions between states in $n, n(q6)$.