ELECTRONIC SUPPLEMENTARY INFORMATION

Nonlinear machine learning and design of reconfigurable digital colloids

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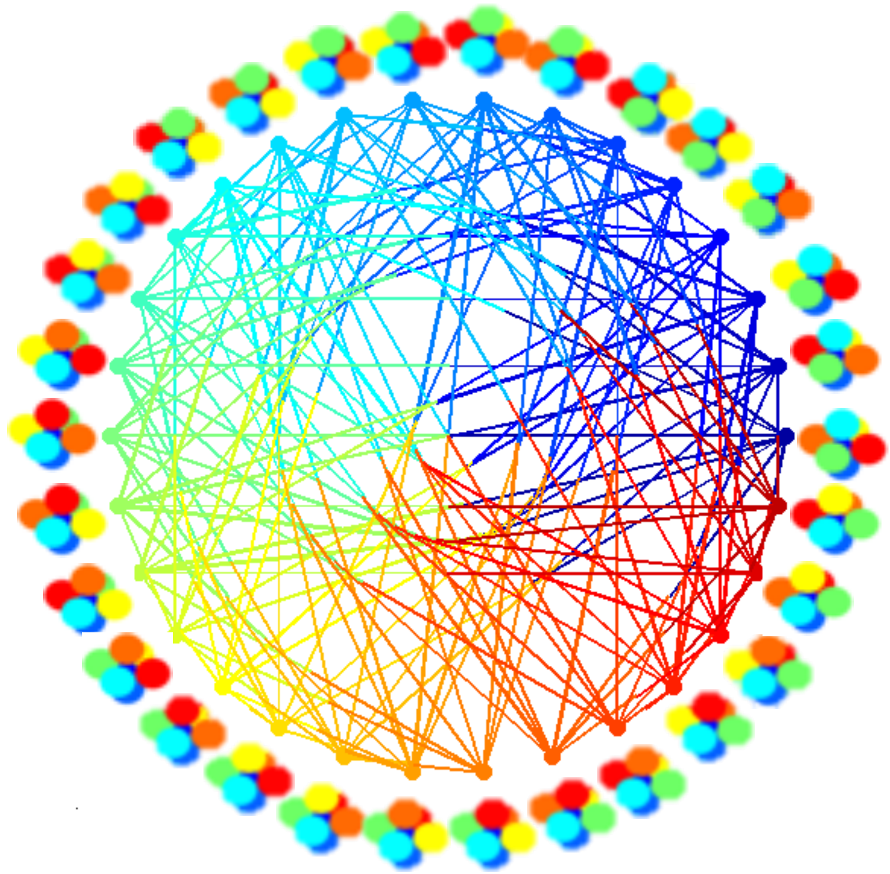
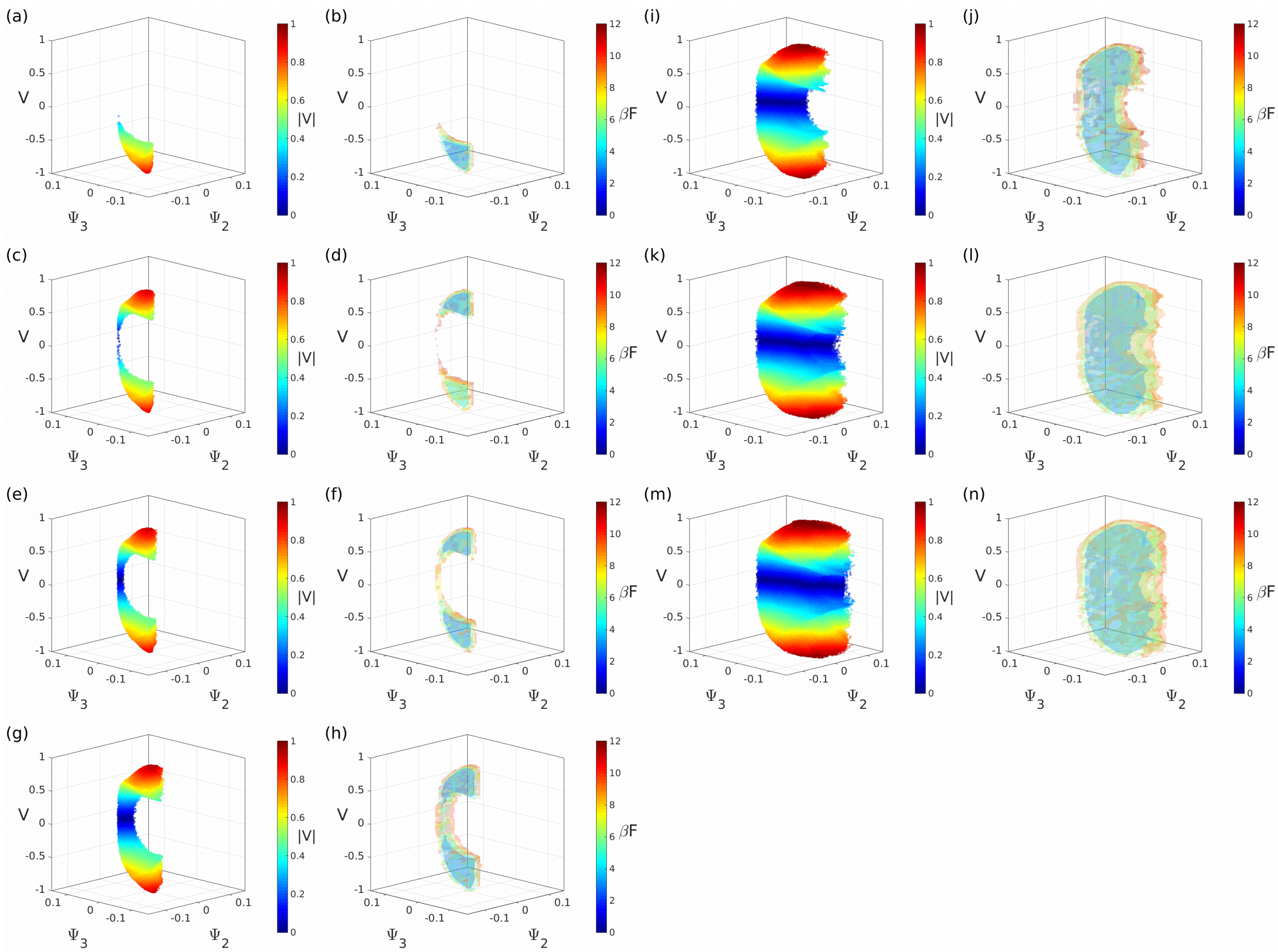
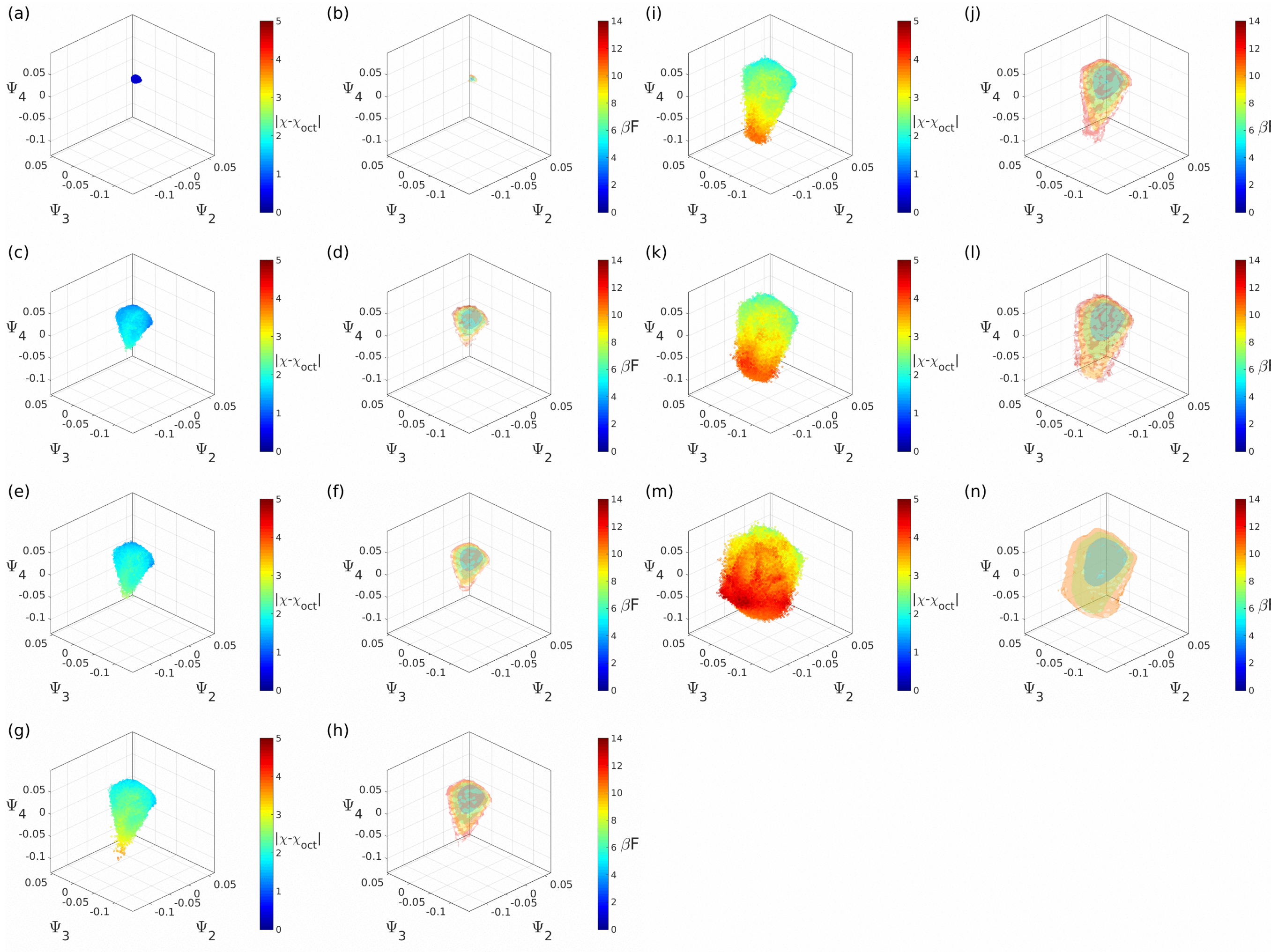


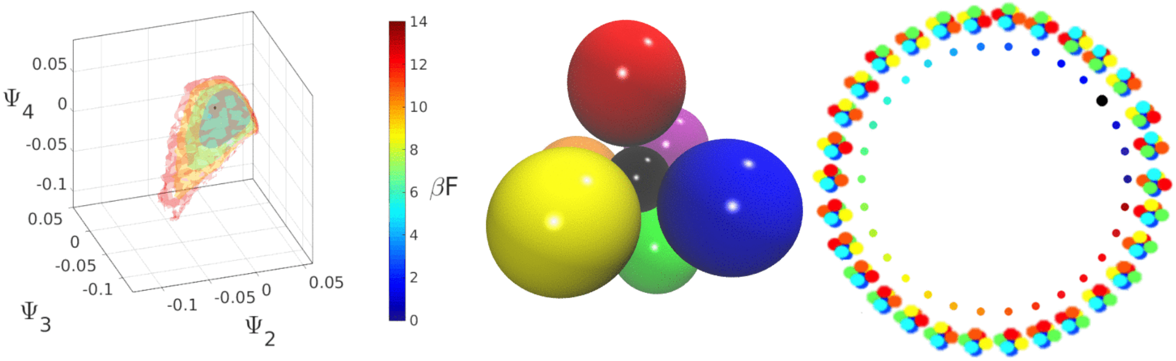
Figure S1: Bit state transition map for the N=6 digital colloid illustrating the 30 rotationally distinguishable bit states as nodes around the exterior of graph and structural transition events linking bit states as edges through the interior. Nodes and edges are color coded to aid in interpretation. Each bit state is able to access eight others through a single halo particle collective transition event. The diameter of the transition network is such that a maximum of three transition events are required to transition between any pair of bit states.



Movie S1: Rotating movies of the diffusion map embeddings and free energy landscapes for the N=4 tetrahedral digital colloid given in Figure 3 of the main text. In each pair of panels we present on the left the low-dimensional projection of the 2×10^6 microstates harvested from the Brownian dynamics simulation at a particular value of Λ into the intrinsic manifold, and on the right the free energy surface over the intrinsic manifold $F(\Psi_2, \Psi_3, V)$. (a,b) $\Lambda = 0.4000$, (c,d) $\Lambda = 0.4142$, (e,f) $\Lambda = 0.4400$, (g,h) $\Lambda = 0.5000$, (i,j) $\Lambda = 0.7500$, (k,l) $\Lambda = 1.0000$, (m,n) $\Lambda = 1.1256$. Points in the diffusion map embeddings are colored by $|V|$. Isosurfaces are plotted at $\beta F = \{0, 2, 4, 6, 8, 10, 12\}$, with the arbitrary zero of free energy defined by the most populated voxel of the embedding.



Movie S2: Rotating movies of the diffusion map embeddings and free energy landscapes for the N=6 octahedral digital colloid given in Figure 6 of the main text. In each pair of panels we present on the left the low-dimensional projection of the 2×10^6 microstates harvested from the Brownian dynamics simulation performed at a particular value of Λ into the intrinsic manifold, and on the right the free energy surface over the intrinsic manifold $F(\Psi_2, \Psi_3, \Psi_4)$. (a,b) $\Lambda = 0.4142$, (c,d) $\Lambda = 0.5100$, (e,f) $\Lambda = 0.5275$, (g,h) $\Lambda = 0.5500$, (i,j) $\Lambda = 0.5750$, (k,l) $\Lambda = 0.6000$, (m,n) $\Lambda = 0.6500$. Points in the diffusion map embedding are colored by the $L_{1,1}$ norm between the normalized halo particle pairwise distances matrix of the corresponding microstate and that of an idealized octahedron $||\chi' - \chi'_{oct}||$. Isosurfaces are plotted at $\beta F = \{0, 2, 4, 6, 8, 10, 12, 14\}$, with the arbitrary zero of free energy defined by the most populated voxel of the embedding.



Movie S3: Tracking a N=6 octahedral digital colloid transition event through the intrinsic manifold and transition network at $\Lambda = 0.5750$. **(middle)** The digital colloid undergoes thermally-driven fluctuations that lead to a collective rearrangement of the halo particles from one of the 30 rotationally distinguishable octahedral bit states to another via a triangular prism transition state. **(left)** We track the structural evolution of the digital colloid as a black point moving over the low-dimensional intrinsic manifold discovered by diffusion maps previously presented in Figure 6j. The digital colloid initially resides in the broad octahedral basin until fluctuations lead to a cooperative transition event wherein the halo particles rearrange into the triangular prism transition structure that ultimately leads to a productive transition to a new rotationally distinguishable bit state. **(right)** The transition event can be visualized as a transformation from one of the 30 rotationally distinguishable octahedral bit states to another as illustrated by the pathway connecting the two nodes in the bit state network. Each bit state is able to access eight others through a single halo particle collective transition event (cf. Figure S1).