Supplementary information for

Thermal-controlled cellular uptake of "hot" nanoparticles

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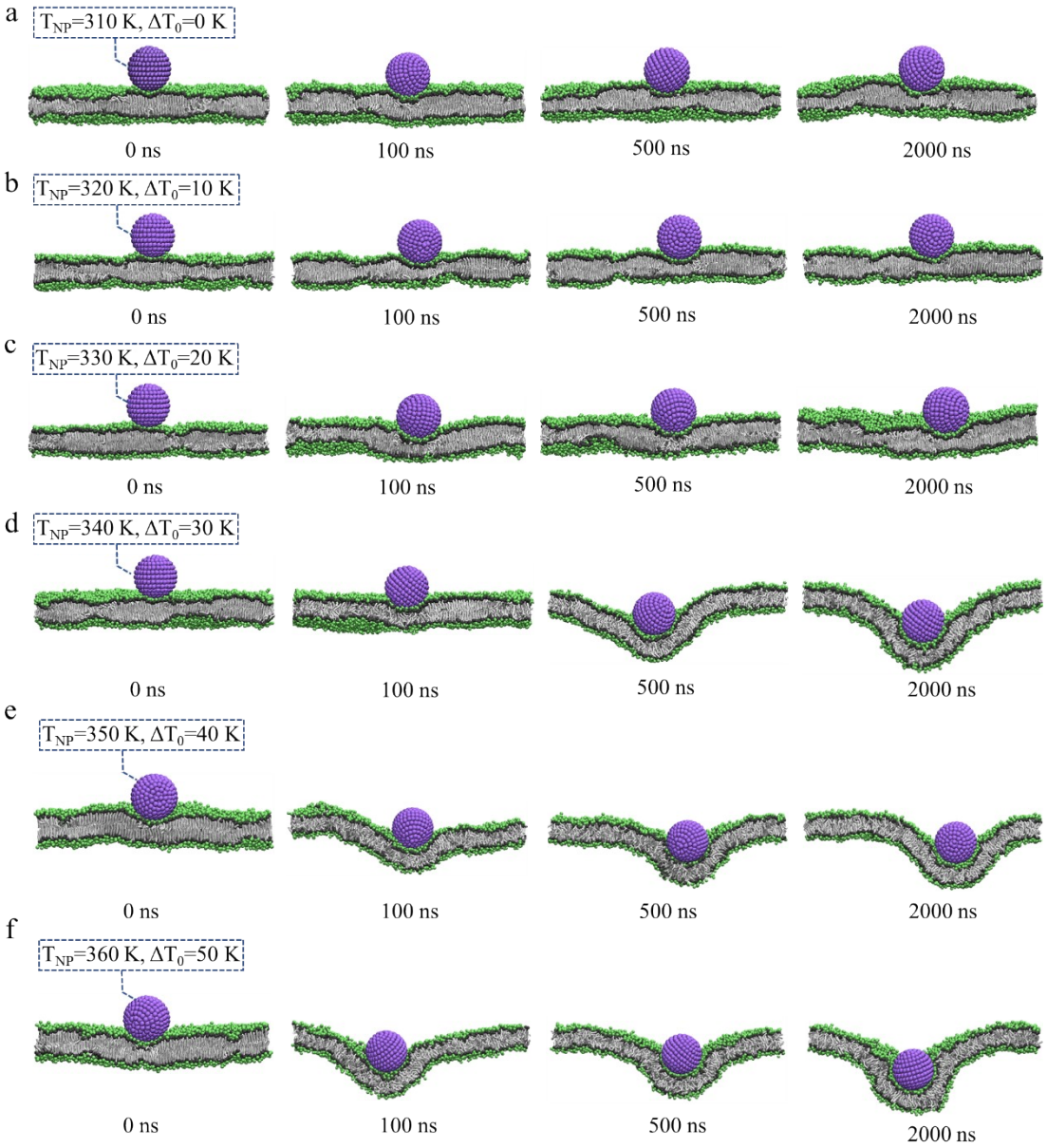

Fig. S1 Representative simulation snapshots showing the NP-membrane interactions with varying T_{NP}. In all cases, $D = 7\ nm$, $\varepsilon = 3.5\ kJ/mol$, $T_{mem}(0) = 310\ K$, membrane lipid type: DPPC, membrane size: 40 × 40 nm^2.

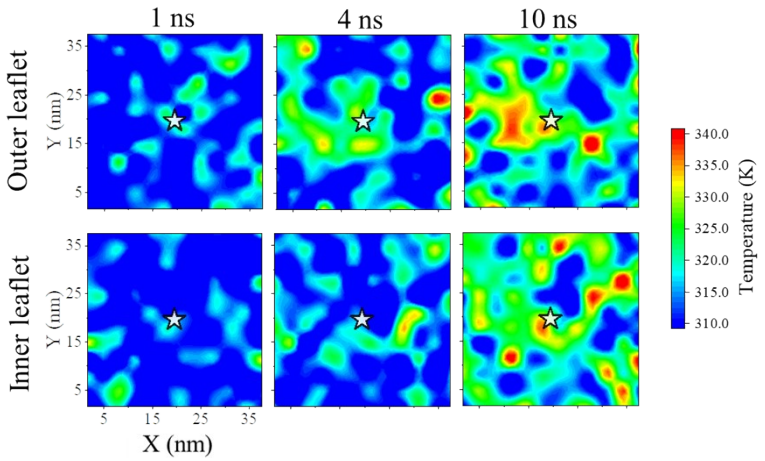

Fig. S2 2D temperature map of lipids (referring to the outer and inner leaflet respectively) at different time points during the NP–membrane interactions. The NP location is marked with stars. $D = 7\ nm$, $\varepsilon = 3.5\ kJ/mol$, $T_{mem}(0) = 310\ K$, $T_{NP} = 340\ K$, membrane lipid type: DPPC, membrane size: 40 × 40 nm^2.

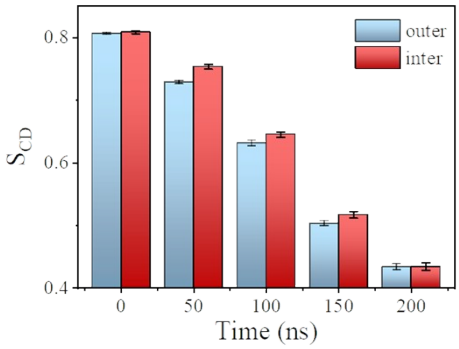

Fig. S3 Changes of the order parameter, S_{cd}, of the two leaflet lipids at the varying time points. In all cases, $D = 7\ nm$, $\varepsilon = 3.5\ kJ/mol$, $T_{mem}(0) = 310\ K$, membrane lipid type: DPPC, membrane size: 80 × 80 nm^2.

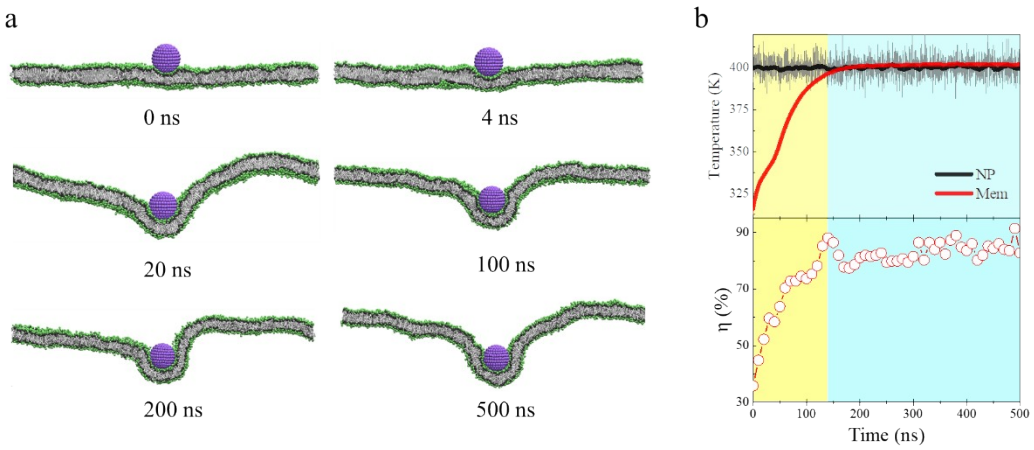

Fig. S4 (a) Representative simulation snapshots showing the interaction between a large bilayer membrane and an irradiated NP with $T_{NP} = 400\ K$. (b) Corresponding time evolution of the component temperature (referring to NP and membrane respectively) and wrapping ratio during the NP–membrane interactions**.** In all cases, $D = 7\ nm$, $\varepsilon = 3.5\ kJ/mol$, $T_{mem}(0) = 310\ K$, membrane lipid type: DPPC, membrane size: 80 × 80 nm^2.

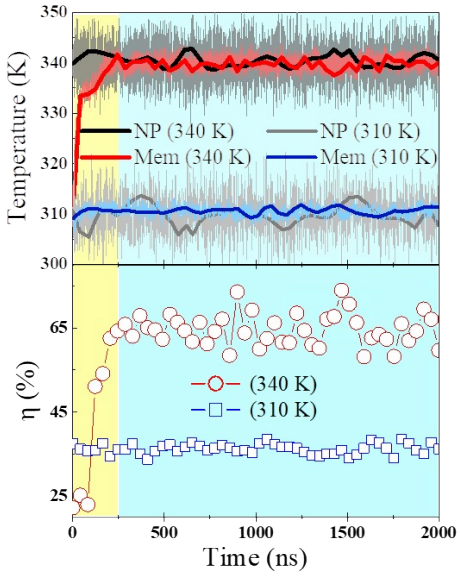

Fig. S5 Evolutions of membrane temperature and the membrane-wrapping ratio of the NP in the heating process**.** The temperature in the brackets refers to T_{NP}. In all cases, $D = 7\ nm$, $\varepsilon = 3.5\ kJ/mol$, $T_{mem}(0) = 310\ K$, membrane lipid type: DPPC, membrane size: 40 × 40 nm^2.

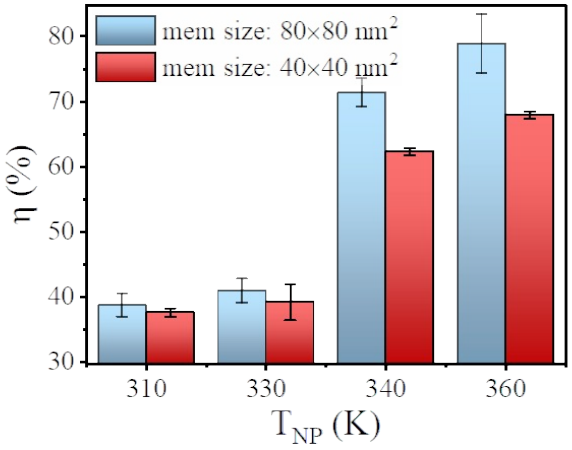

Fig. S6 Comparison of the dependence of membrane-wrapping ratio of NPs, η, on T_{NP} in the two simulation systems with varying membrane sizes. In all cases, $D = 7\ nm$, $\varepsilon = 3.5\ kJ/mol$, $T_{mem}(0) = 310\ K$, membrane lipid type: DPPC.

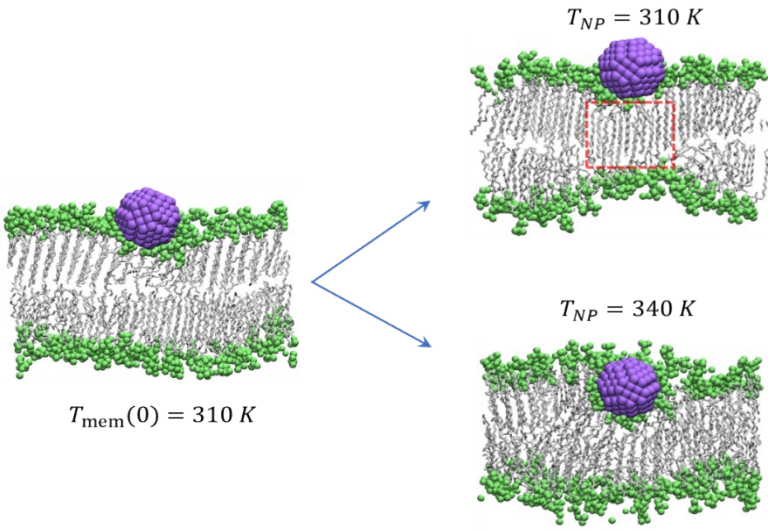

Fig. S7 Representative all-atom simulation snapshots showing the interaction between a DPPC lipid bilayer membrane and an irradiated NP with varying T_{NP}. The dashed red box highlights the interdigitation of lipid tails. In all cases, $D = 2\ nm$, $T_{mem}(0) = 310\ K$, membrane lipid type: DPPC, membrane size: 10 × 10 nm^2.

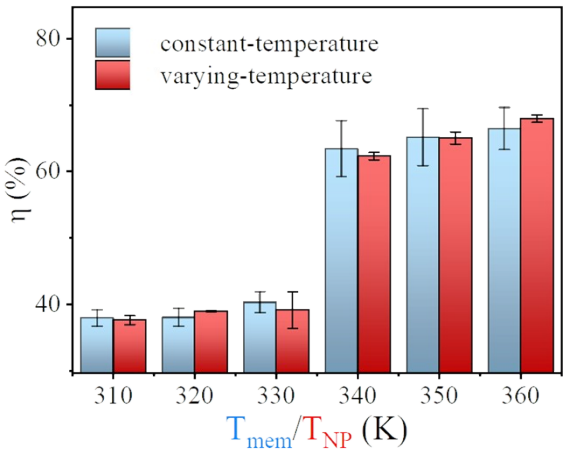

Fig. S8 Histograms showing the membrane-wrapping ratio of NPs, η, with different T_{NP} or T_{mem} and simulation conditions. In all cases, $D = 7\ nm$, $\varepsilon = 3.5\ kJ/mol$, membrane lipid type: DPPC. For the varying-temperature simulations (i.e., heating), $T_{mem}(0) = 310\ K$, membrane size: 40 × 40 nm^2.

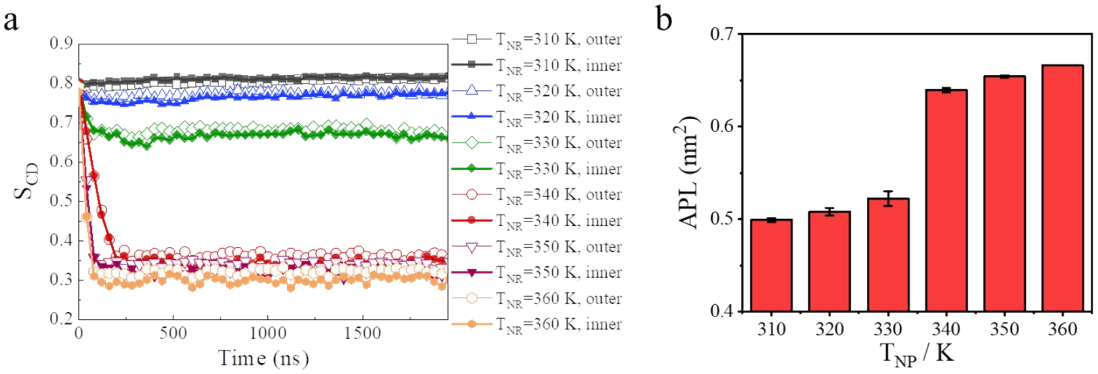

Fig. S9 (a) Evolution of the order parameter, S_{cd}, of lipids. (b) Change of APL with T_{NP}. In all cases, $D = 7\ nm$, $\varepsilon = 3.5\ kJ/mol$, $T_{mem}(0) = 310\ K$, membrane lipid type: DPPC.

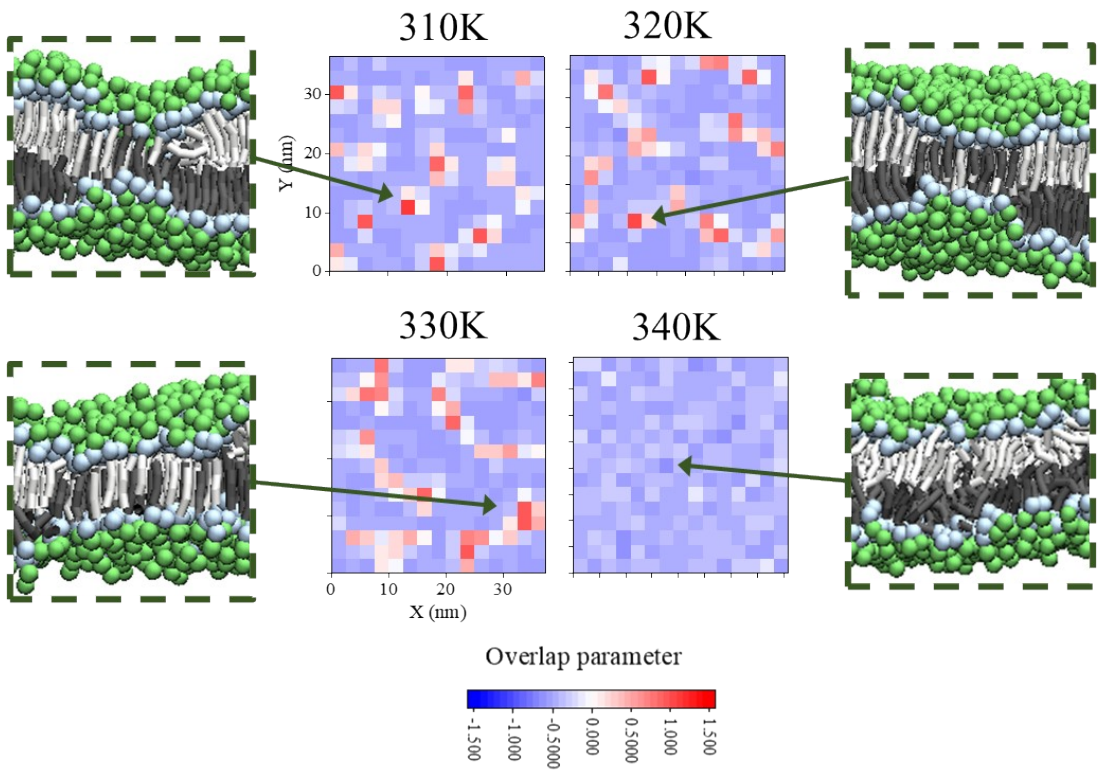

Fig. S10 2D topographic map of the overlap parameter ϕ. All the data points were averaged based on the last 250 ns trajectories. Representative snapshots were shown correspondingly. The tails of lipids in the inner and outer leaflets are colored in dark gray and silver, respectively.

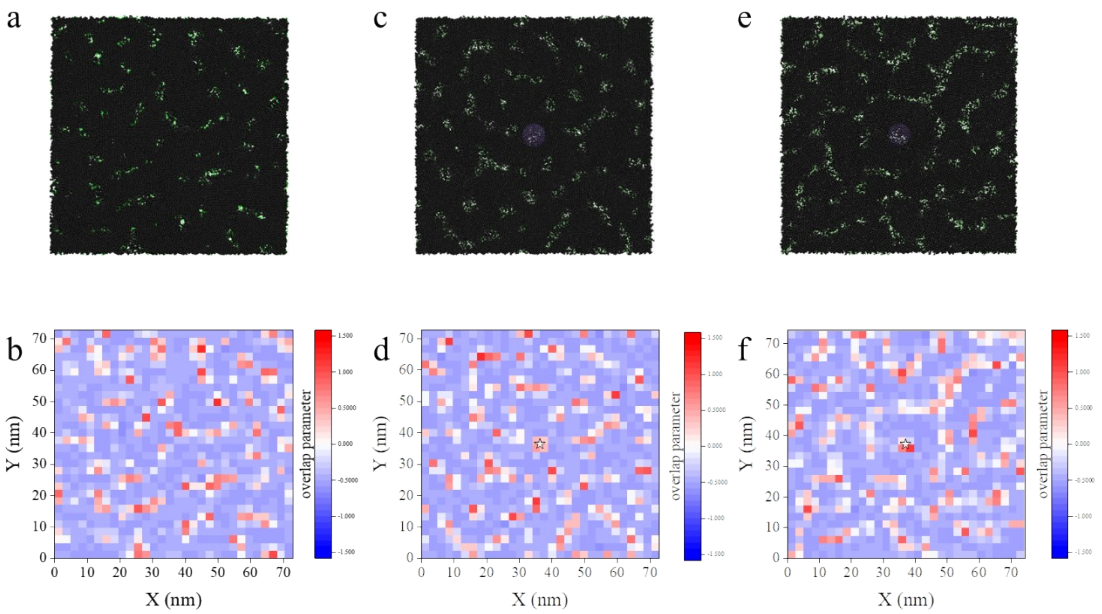

Fig. S11 Representative simulation snapshots showing the formation of lipid interdigitation in the large membrane system. (a, b) without the NP. $T_{mem}(0) = 310\ K$; (c, d) with a NP. $T_{mem}(0) = 310\ K$, $T_{NP} = 310\ K$; (e, f) with a NP. $T_{mem}(0) = 310\ K$, $T_{NP} = 320\ K$. $D = 7\ nm$, $\varepsilon = 3.5\ kJ/mol$, membrane lipid type: DPPC, membrane size: 80 × 80 nm^2. Green: DPPC head; Black: DPPC tail. Only inner leaflet is shown for clarification, and light-colored regions are those with lipid interdigitation.

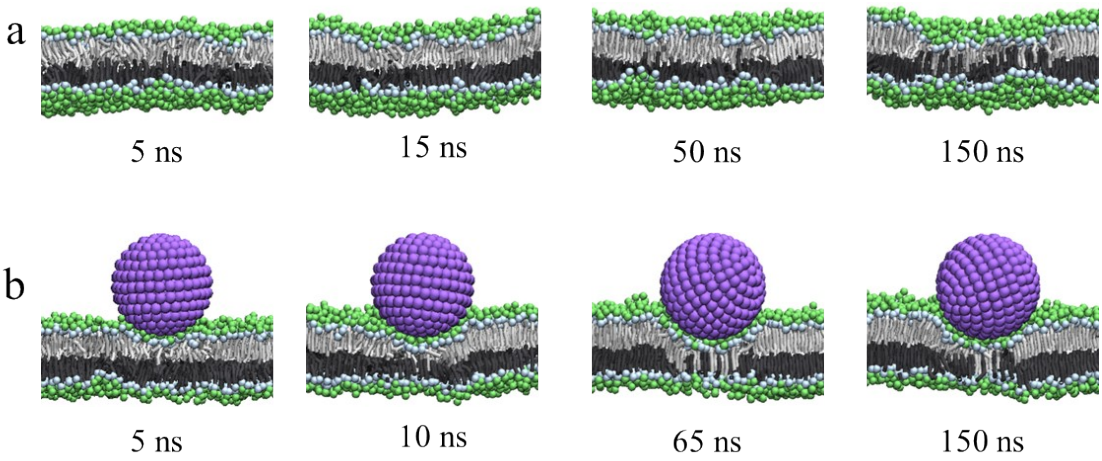

Fig. S12 Representative simulation snapshots showing the formation of lipid interdigitation in the membrane. (a) the region away from the NP; (b) the region near the NP. $T_{mem}(0) = 310\ K$, $T_{NP} = 320\ K$. $D = 7\ nm$, $\varepsilon = 3.5\ kJ/mol$, membrane lipid type: DPPC, membrane size: 80 × 80 nm^2. The tails of lipids in the inner and outer leaflets are colored in dark gray and silver, respectively.

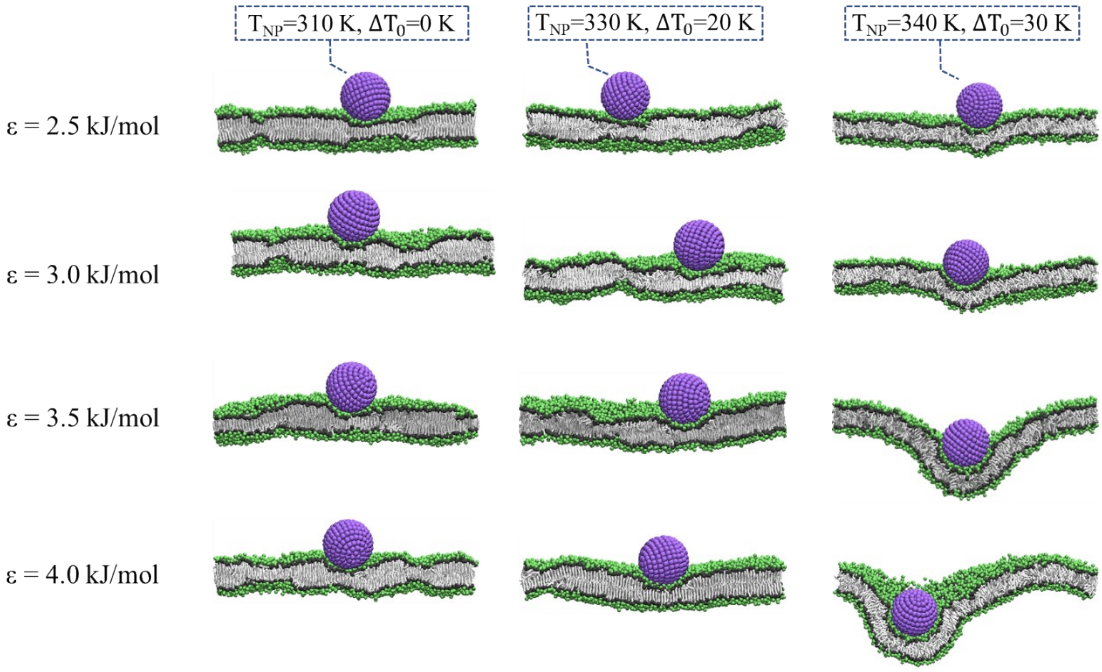

Fig. S13 Representative simulation snapshots showing the interaction between NP and DPPC lipid bilayer membrane with different T_{NP} and ε. In all cases, $D = 7\ nm$, $T_{mem}(0) = 310\ K$, membrane lipid type: DPPC, membrane size: 40 × 40 nm^2.

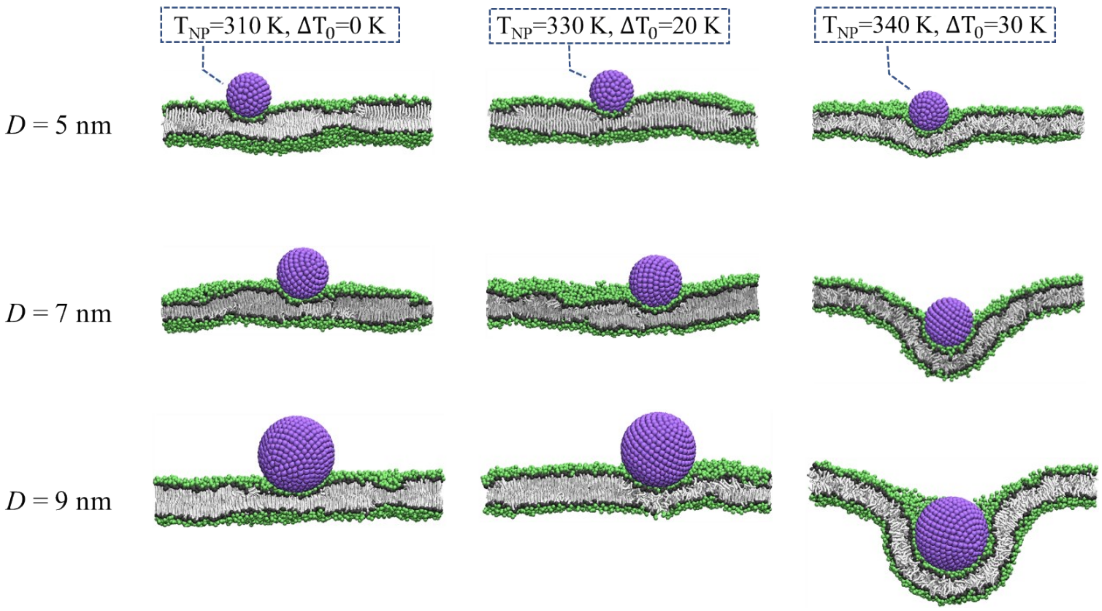

Fig. S14 Representative simulation snapshots showing the interaction between NP and DPPC lipid bilayer membrane with different T_{NP} and D. In all cases, $\varepsilon = 3.5\ kJ/mol$, $T_{mem}(0) = 310\ K$, membrane lipid type: DPPC, membrane size: 40 × 40 nm^2.

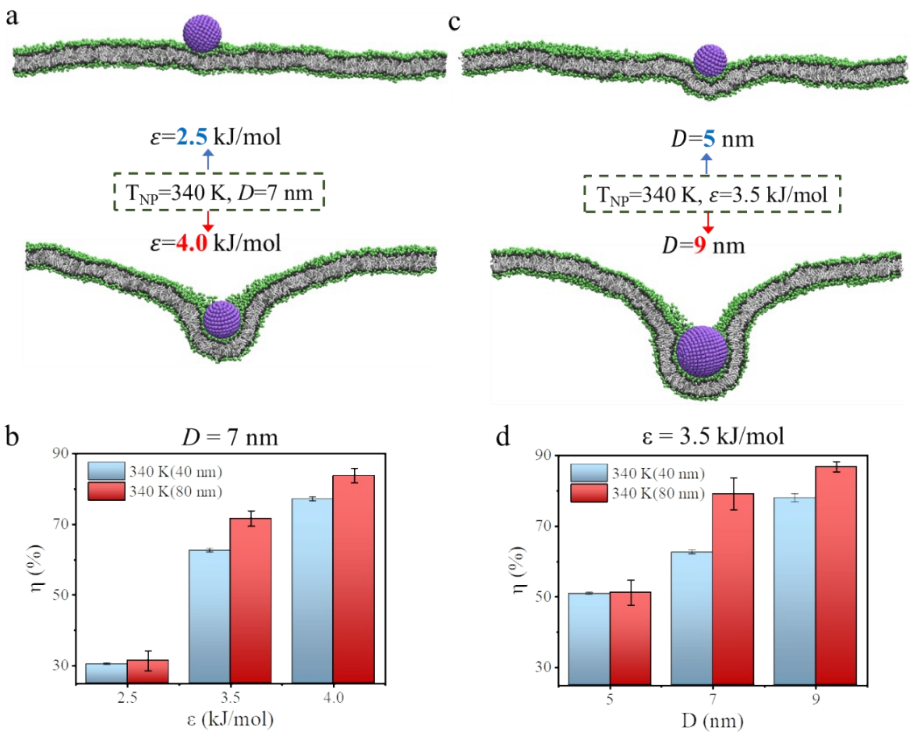

Fig. S15 (a, b) Influence of NP-membrane affinity ε on the membrane-wrapping effect in the large membrane system. (c, d) Influence of NP size D on the membrane-wrapping effect in the large membrane system. In all cases, $T_{NP} = 340\ K$, $T_{mem}(0) = 310\ K$, membrane lipid type: DPPC, membrane size: 80 × 80 nm^2.

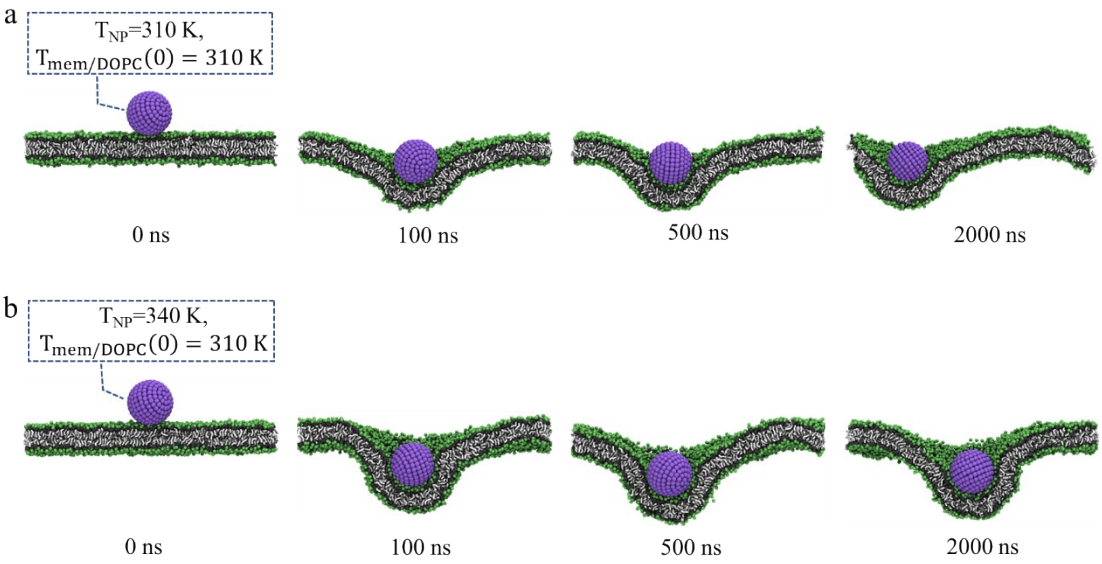

Fig. S16 Representative simulation snapshots showing the interaction between NP and a DOPC lipid bilayer membrane with different T_{NP}. In all cases, $D = 7\ nm$, $\varepsilon = 3.5\ kJ/mol$, $T_{mem}(0) = 310\ K$, membrane lipid type: DOPC, membrane size: 40 × 40 nm^2.

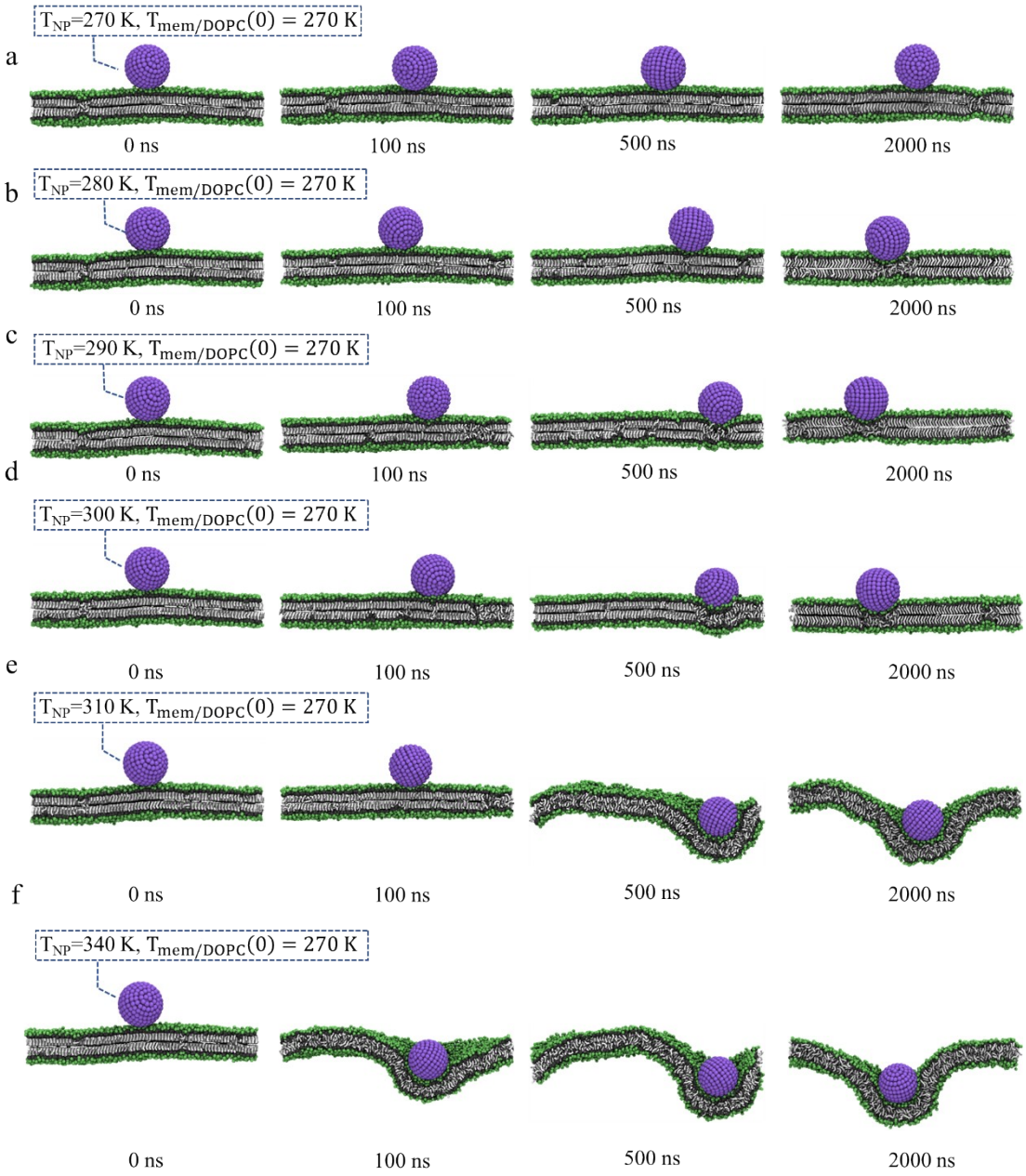

Fig. S17 Representative simulation snapshots showing the interaction between NP and a DOPC lipid bilayer membrane with different T_{NP}. In all cases, $D = 7\ nm$, $\varepsilon = 3.5\ kJ/mol$, $T_{mem}(0) = 270\ K$, membrane lipid type: DOPC, membrane size: 40 × 40 nm^2.

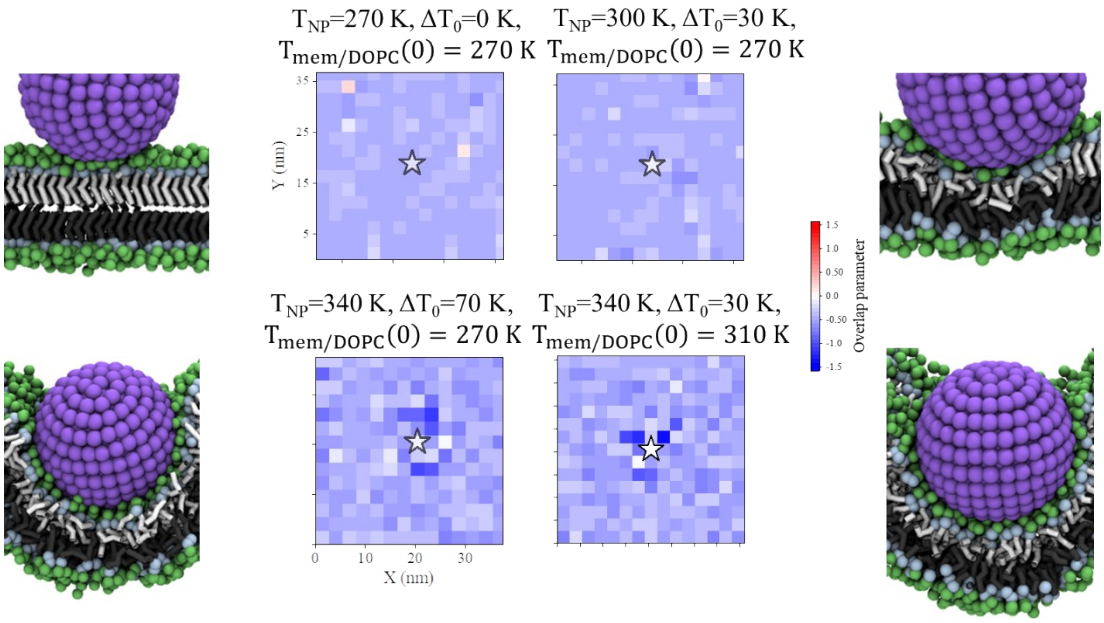

Fig. S18 2D topographic map of the overlap parameter ϕ. The location of NP is marked with stars. For convenience of observation and comparison, the system is translated to keep the location of NP at the center. All the data points were averaged based on the last 250 ns trajectories. Representative snapshots were shown correspondingly. The tails of lipids in the inner and outer leaflets are colored in dark gray and white, respectively. In all cases, $D = 7\ nm$, $\varepsilon = 3.5\ kJ/mol$, $T_{mem}(0) = 310\ K$, membrane lipid type: DOPC, membrane size: 40 × 40 nm^2.