Electronic Supplementary Material (ESI) for Soft Matter.

Electronic Supplementary Information

Directed rolling of positively charged nanoparticles along a flexibility gradient of long DNA molecules

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ESI Contents

Figure S1. Persistence length of DNA models

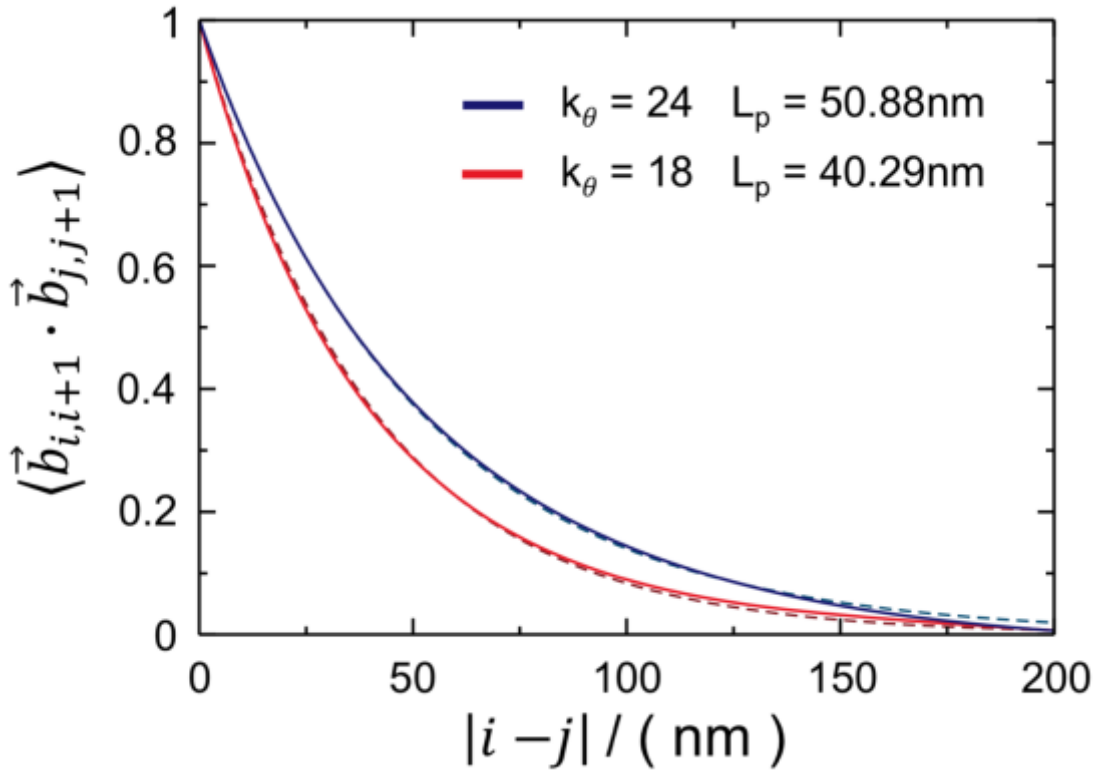

Figure S1. Calculation of the persistence lengths of DNA models with uniform flexibility of $k_\theta = 18k_BT/rad^2$ and $24k_BT/rad^2$ in terms of the average correlation function between DNA bond vectors as described in Eqn. (4) of the main text. The persistence length of a DNA model is 40.3 nm and 50.9 nm when $k_\theta = 18k_BT/rad^2$ and $24k_BT/rad^2$.

Figure S2. Distribution of the number of DNA monomers in contact with a nanoparticle

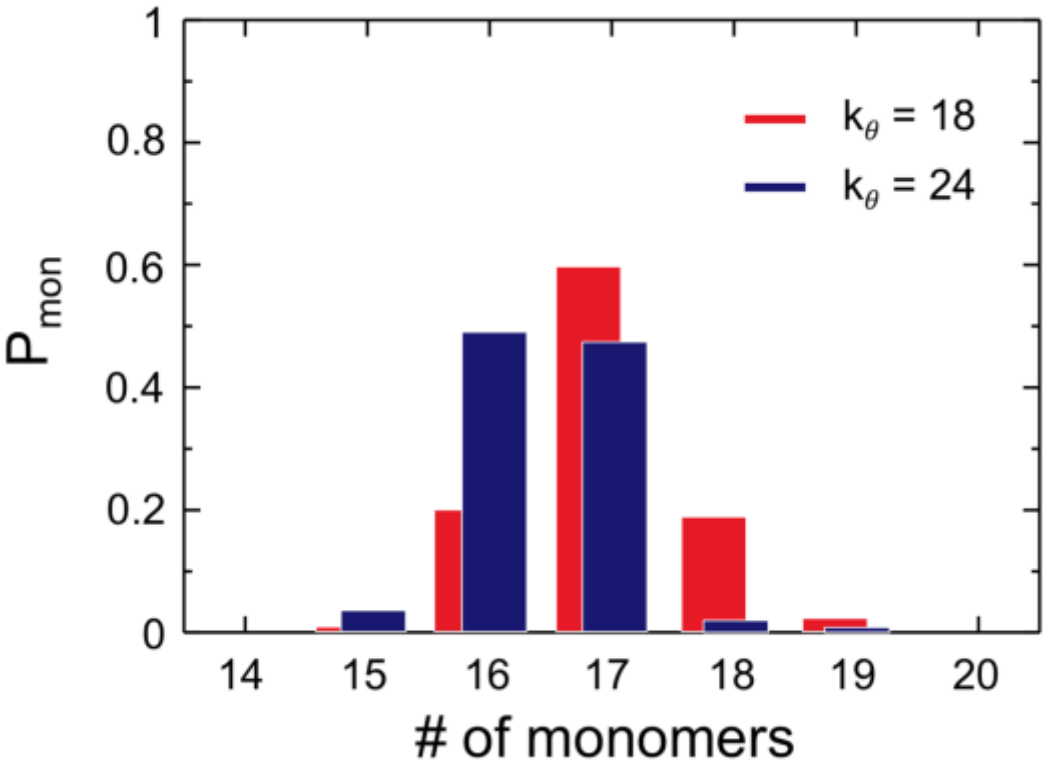

Figure S2. Distribution of the number of DNA monomers wrapping a nanoparticle for two DNA models of $k_\theta = 18k_BT/rad^2$ and $24k_BT/rad^2$. Monomers are considered in contact with a nanoparticle when they are within the distance of $(\sigma_{mon} + \sigma_{NP})/2 \times 1.2 = 1.8\sigma$ from a nanoparticle, where σ_{mon} and σ_{NP} are the diameters of a DNA monomer and a nanoparticle, respectively. The average numbers of the contacting DNA monomers are 17.02 ± 0.01 and 16.47 ± 0.01 for $k_\theta = 18k_BT/rad^2$ and $24k_BT/rad^2$.

Figure S3. Three patterns of DNA flexibility gradients

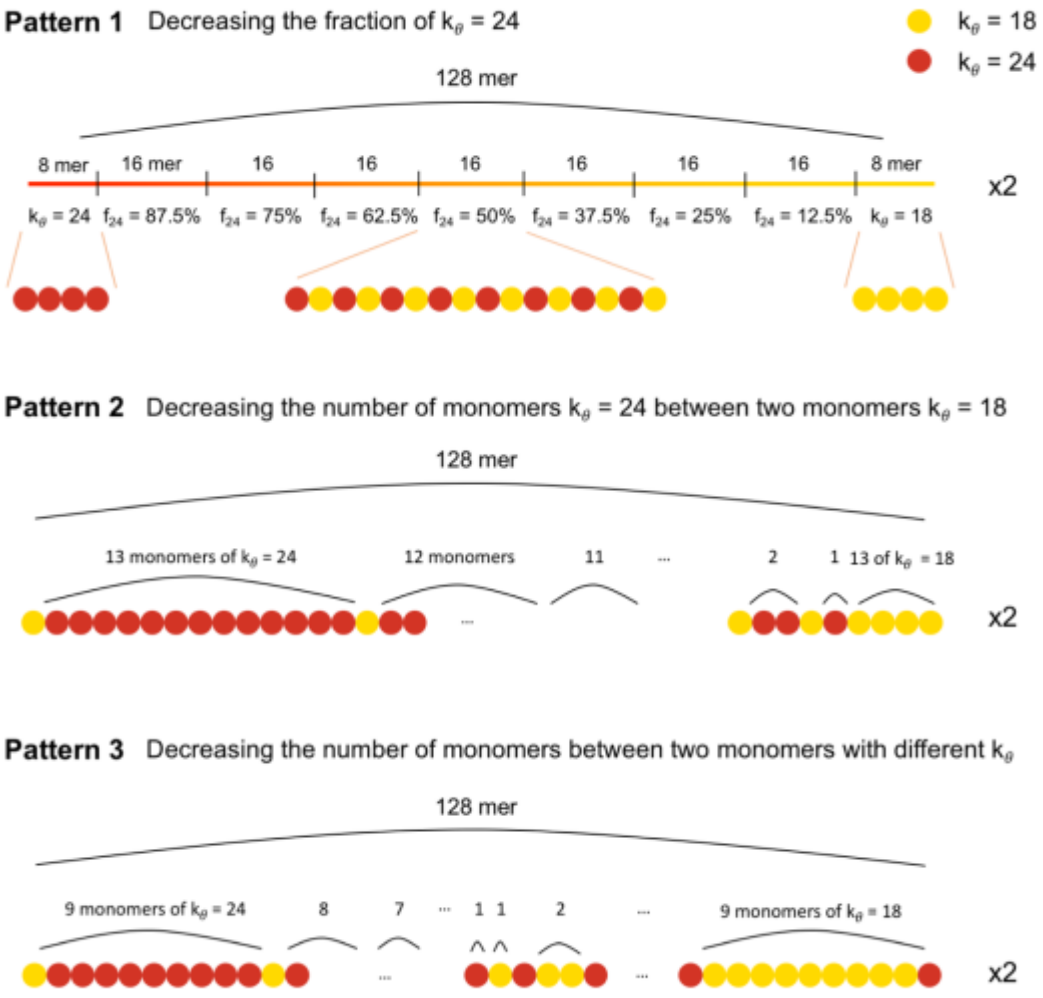

Figure S3. Three patterns of DNA flexibility gradients, among which Pattern 1 is presented in Figure 2 of the main text. DNA monomers with different colors indicate that the monomer and next two monomers make an angle that is subject to a harmonic angle restraint with bending parameters of $k_\theta = 18k_BT/rad^2$ (yellow) and $24k_BT/rad^2$ (red).

Figure S4. Directed rolling of a nanoparticle on DNA flexibility gradients

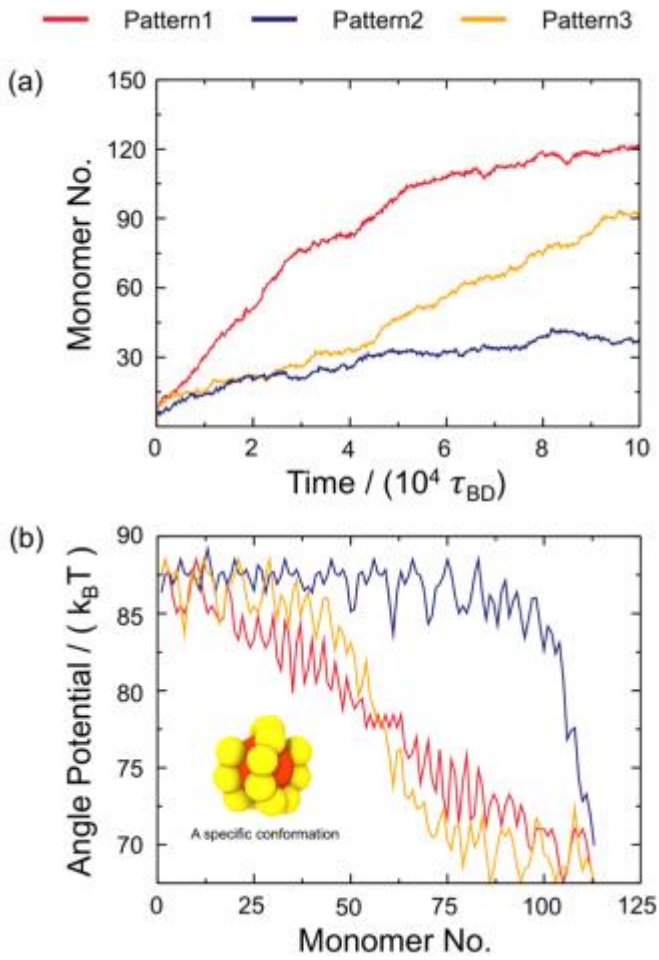

Figure S4. a) Average trajectory of a nanoparticle along three patterns of DNA flexibility gradients depicted in Figure S3 obtained from 20 independent simulations. (b) Angle potential energy, calculated for a specific conformation shown in the figure, along flexibility gradients assuming that the conformation is maintained during rolling. The angle potential energy along the flexibility gradient is approximated as the variation of thermodynamic stability of DNA-nanoparticle binding, suggesting the efficiency of directional rolling of a nanoparticle. The results for Pattern 1 are presented in Figure 6 of the main text.

Figure S5. Nanoparticle binding with electric charges +32 and +16

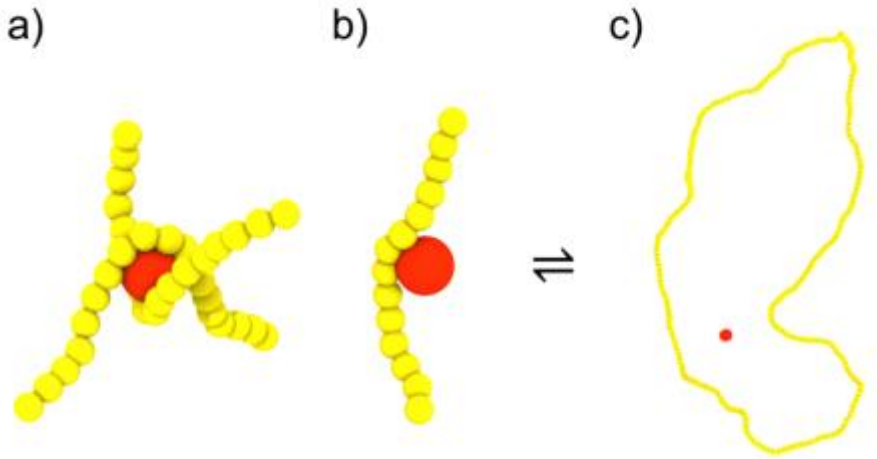

Figure S5. Snapshots from the simulations of a nanoparticle with charge (a) +32 and (b) and (c) +16. (a) A nanoparticle with charge +32 often forms a bridging conformation by connecting different regions of the same ring DNA molecule. (b) and (c) A nanoparticle with charge +16 repeats the binding to and dissociation from a DNA molecule. The observation of stepwise rolling dynamics along DNA is difficult for nanoparticles with charge +32 and +16,

Figure S6. 20 independent trajectories of a nanoparticle with charge +64 and +48 on a DNA flexibility gradient

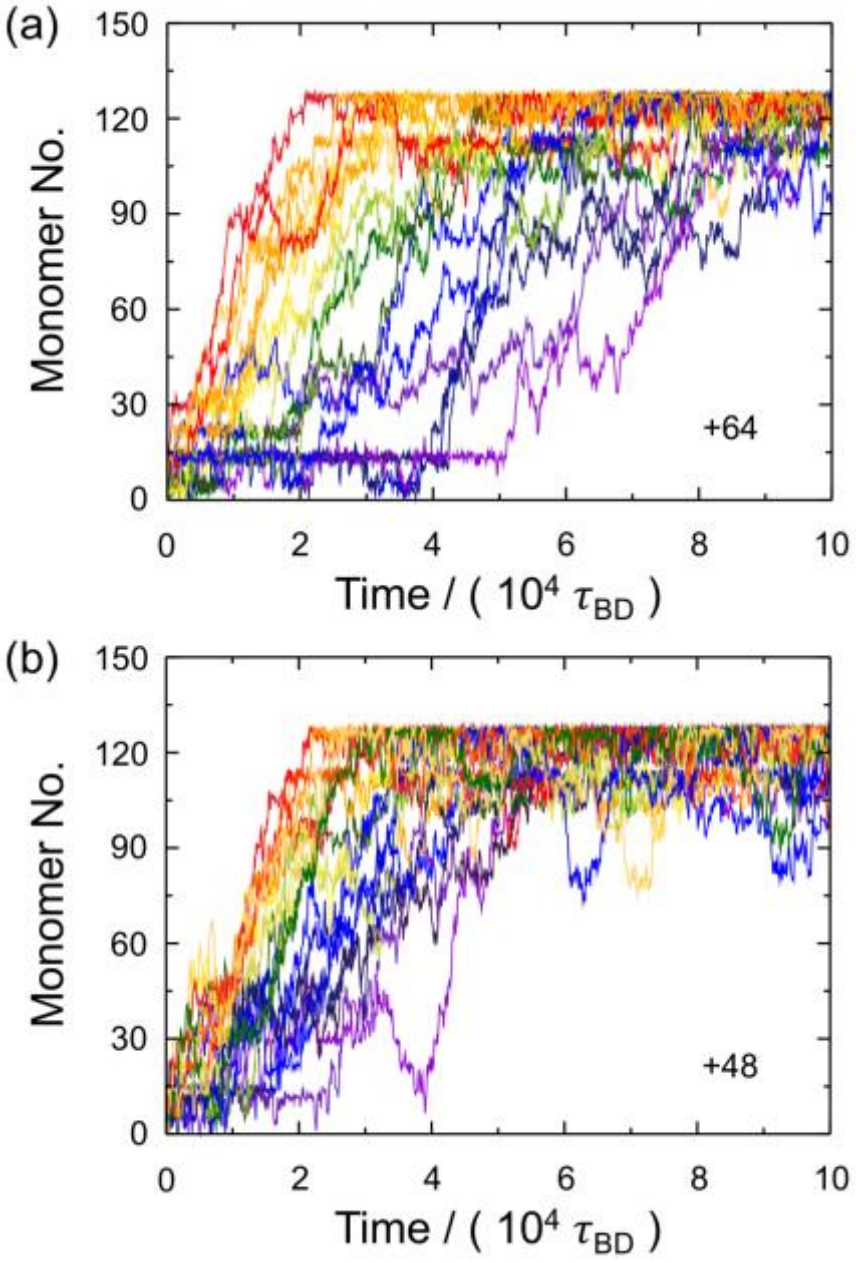

Figure S6. All 20 independent trajectories of a nanoparticle with charge (a) +64 and (b) +48 along a DNA flexibility gradient (Pattern 1). The averages are compared in Figure 7 (b) of the main text.

Figure S7. Nanoparticle binding at different ion concentrations

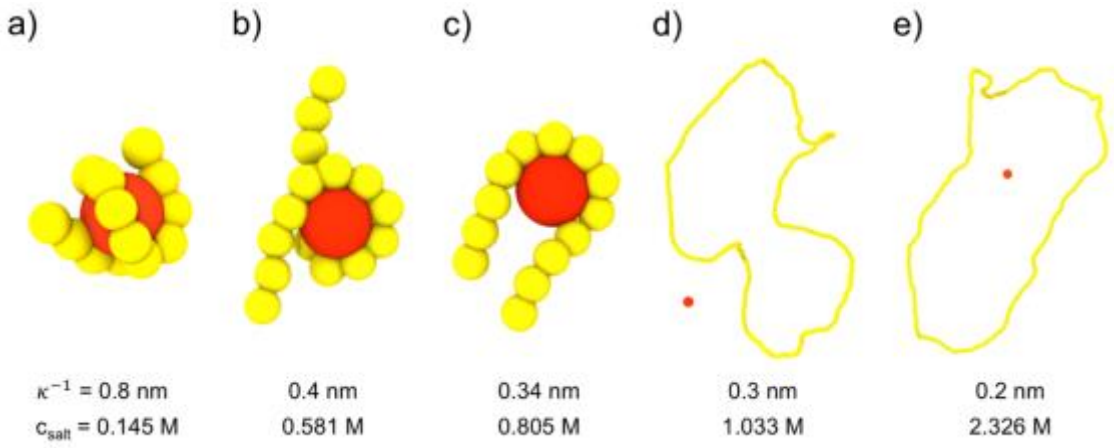

Figure S7. Snapshots from simulations of a nanoparticle of charge +64 bound to a ring DNA molecule in various salt concentrations. (a) The Debye length (κ^{-1}) is 0.8 nm and the salt concentration (c_{salt}) is 0.145 M. These parameters are used for all the results presented in the main text. (b) $\kappa^{-1} = 0.4$ nm and $c_{salt} = 0.581$ M. (c) $\kappa^{-1} = 0.34$ nm and $c_{salt} = 0.805$ M. (d) $\kappa^{-1} = 0.3$ nm and $c_{salt} = 1.033$ M. (e) $\kappa^{-1} = 0.2$ nm and $c_{salt} = 2.326$M. At high salt concentrations the electrostatic interaction of nanoparticles are screened and they do not remain bound to DNA.