

Supporting information for:

**How do non-Covalent Complexes Dissociate in
Droplets? A case study of the Desolvation of dsDNA
from a Charged Aqueous Nanodrop**

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MD Movies

Movie S1. Denaturation of the 11-mer all A·T dsDNA in a $-20 e$ charged aqueous droplet during desolvation. (150 ps)

File: [mov-dsDNA-denature-gas-phase-150ps.mpg](#)

Movie S2. Denaturation of the 11-mer all A·T DNA duplex in a $-15 e$ charged aqueous droplet that contains 5 Na^+ ions during desolvation. (250 ps)

File: [mov-dsDNA-denature-5Na-gas-phase-250ps.mpg](#)

Movie S3. Ejection of a Cl^- from a droplet composed of 880 H_2O molecules, the 11-mer all A·T dsDNA, 20 Na^+ ions, and 10 Cl^- ions during desolvation into the gas phase. (150 ps)

File: [mov-dsDNA-denature-20Na-10Cl-Cl-ejection-150ps.mpg](#)

Desolvated states of $[\text{dsDNA}]^{20-}$

In Fig. S1 typical snapshots of the desolvated states of $[\text{dsDNA}]^{20-}$ at various ion concentrations are shown. In Fig. S1 (a) and (b) the $[\text{dsDNA}]^{20-}$ denatures when the net charge is more negative than half of the charge of the $[\text{dsDNA}]^{20-}$. (c) The zipped-up double-stranded state of the DNA is formed when the net charge is exactly half that of the $[\text{dsDNA}]^{20-}$. (d)–(f) The compactness of the dsDNA increases when sodium chloride aggregates are formed on the dsDNA.

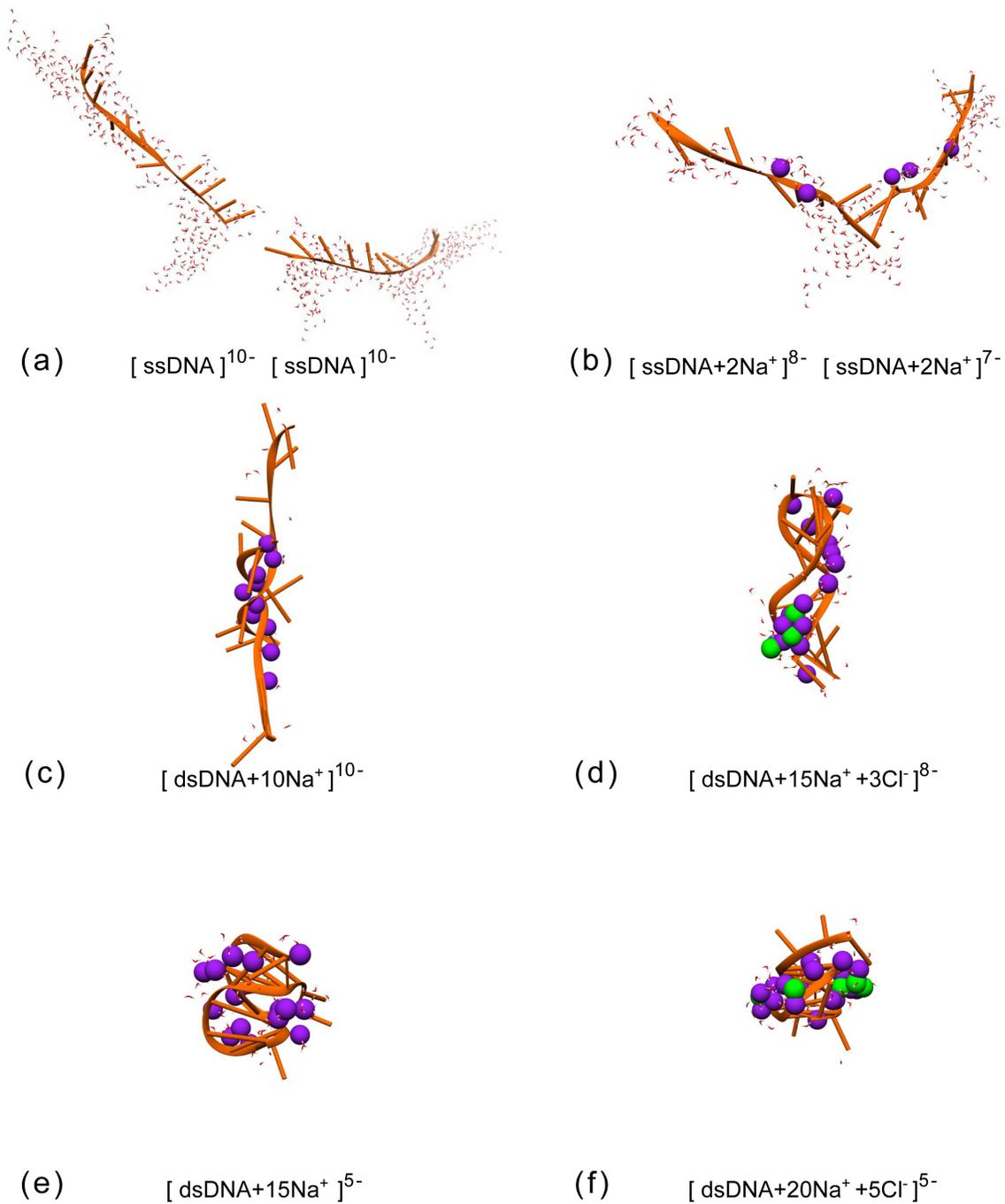


Figure S1: Typical snapshots of the final charge states of dsDNA in the the gas phase. The colour coding is: Na^+ ions are shown by purple spheres and Cl^- ions by green spheres. Details are described in the text.

Parameters of the simulations

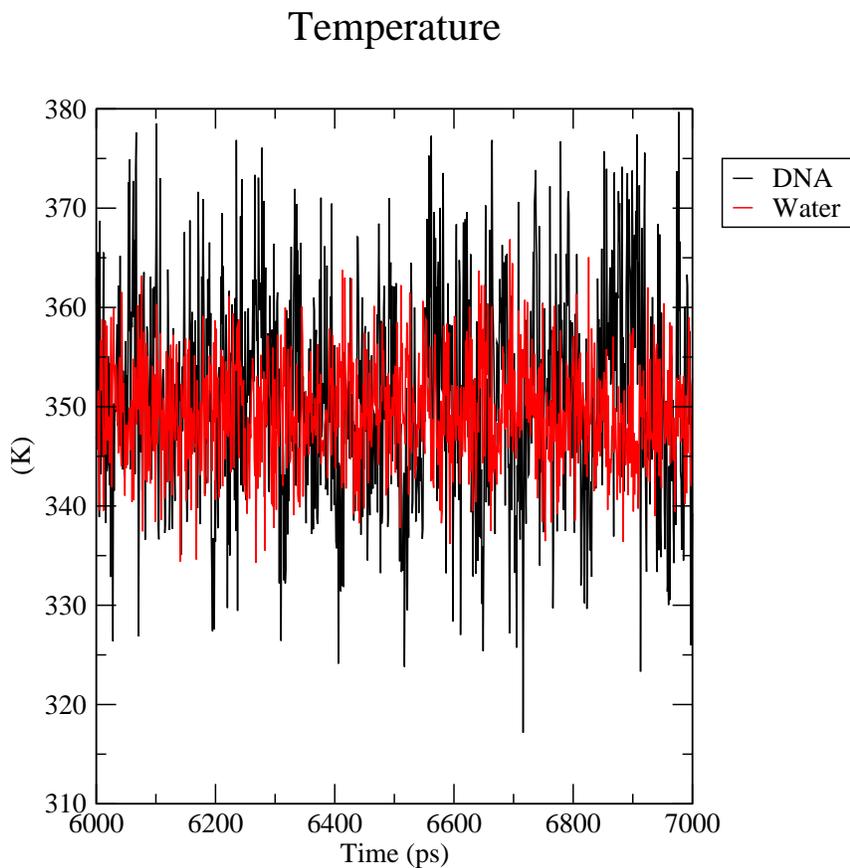


Figure S2: Temperature profile of water and DNA. The droplet comprised ~ 800 TIP3P water molecules and a $[\text{dsDNA}]^{20-}$. The system was coupled to Nosé-Hoover thermostat at $T = 350$ K.

A typical time step for simulations performed with empirical force fields is 1 fs. In our simulations, we use the time step of 0.5 fs, which is 13 times smaller than the fastest O–H vibration in the system. To confirm the choice of our time step we performed short simulations with a time step of 1 fs and we found similar results to that with 0.5 fs regarding the critical charge of the droplet as shown in Fig. S3 with respect to the charge state of the droplet. However, we cannot use 1 fs in our simulations because it is a large time step to resolve the motions in our system. Since, we have evaporation with molecules leaving the droplets in high speed, a larger time step than 0.5 fs may

render the integration algorithm unstable. We also tested the outcome of the fragmentation using the SPC/E model of water. The results are presented in Fig. S3.

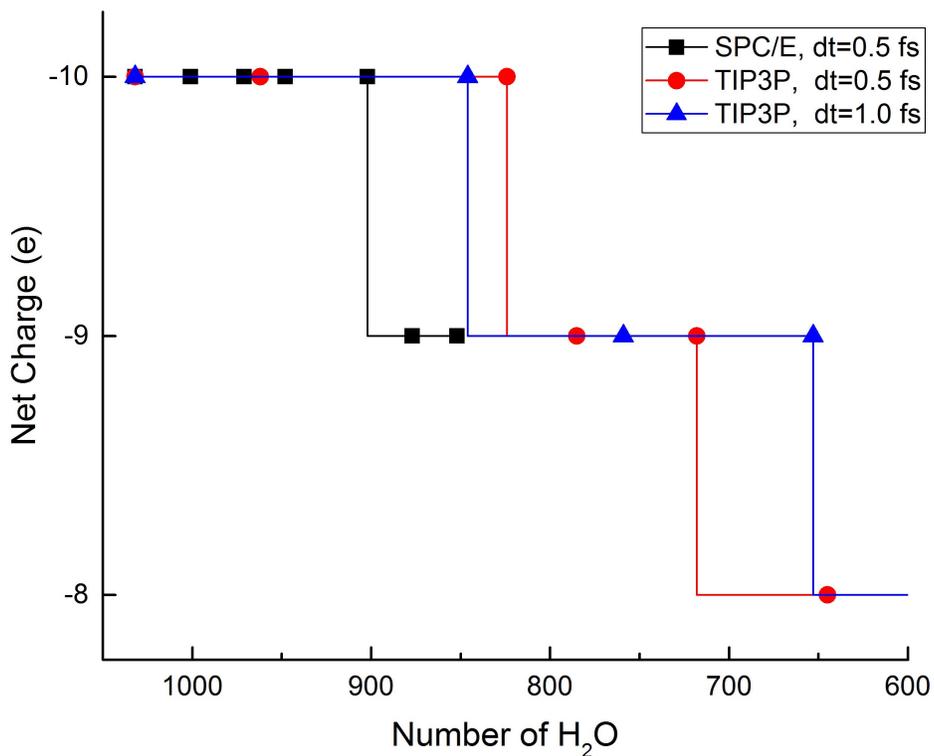


Figure S3: Critical charge of TIP3P and SPC/E water models. Each symbol indicates the net charge of the nanodrop prior to a single Cl⁻ ion evaporation. The change in the charge is illustrated by a vertical line from the data point followed by a horizontal line that indicates the water evaporation before reaching a new critical charge.