Supporting Information: Improved Model of Hydrated Calcium Ion for Molecular Dynamics Simulations Using Classical Biomolecular Force Fields

Jejoong Yoo,[†,‡] James Wilson,[†] and Aleksei Aksimentiev*[,†,‡,¶]

†Department of Physics, University of Illinois at Urbana–Champaign, 1110 West Green Street, Urbana, Illinois 61801

‡Center for the Physics of Living Cells

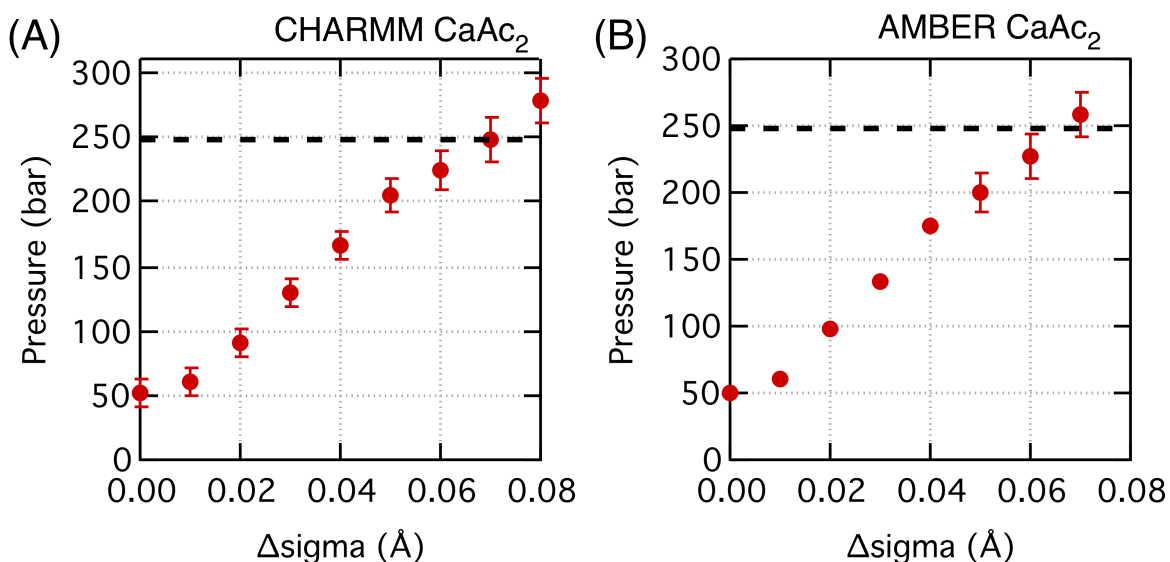¶Beckman Institute for Advanced Science and Technology

E-mail: aksiment@illinois.edu

Figure S1: Simulated osmotic pressure of a 2 m $CaAc_2$ solution (red marks) as a function of the correction to the LJ σ parameter describing the interactions between oxygen atoms of $Ca^{2+}(H_2O)_7$ and oxygen atoms of Ac^- for CHARMM (A) and AMBER (B) models of Ac^-. Error bars indicate the standard deviation of 1 ns block averages of osmotic pressure. In both panels, the black dashed lines depict the experimental osmotic pressure of the 2 m $CaAc_2$ solution at room temperature.[1]
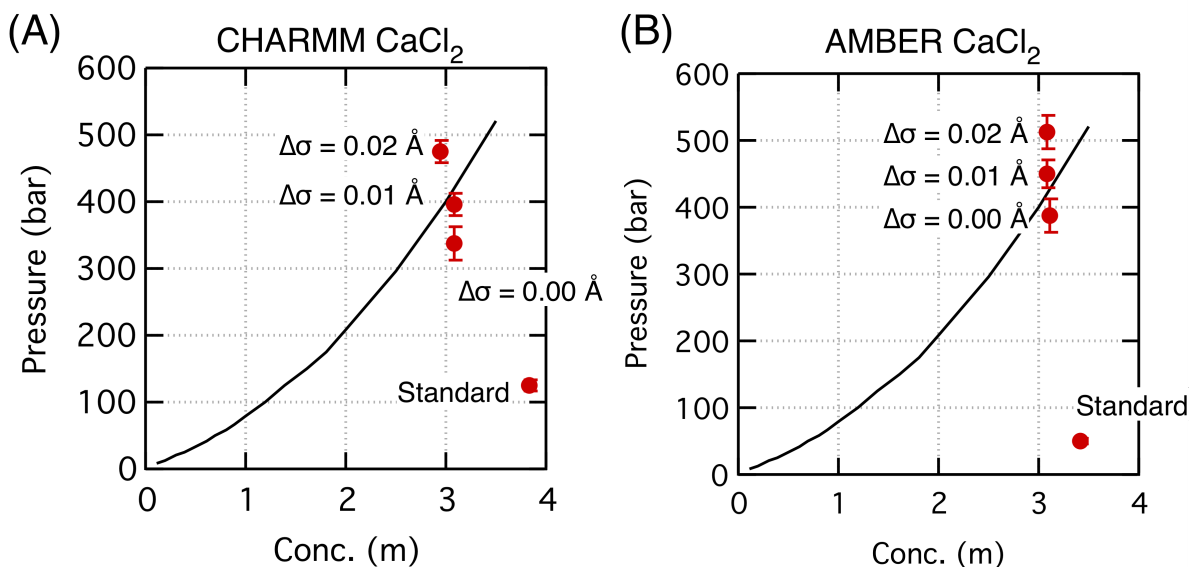
Figure S2: Simulated osmotic pressure of a ~ 3 m $CaCl_2$ solution obtained using the CHARMM (A) and AMBER (B) models of Cl^-. The simulated values are shown as red symbols. "Standard" data points indicate the osmotic pressure values obtained using the standard variants of the force fields. All other data points derived from the simulations performed using our $Ca^{2+}(H_2O)_7$ model and differ from one another by the value of the correction to the LJ σ parameter describing the interactions between oxygen atoms of $Ca^{2+}(H_2O)_7$ and Cl^-; the correction value is listed near each data point. Error bars indicate the standard deviation of 1 ns block averages of osmotic pressure. In both panels, the black line depicts the experimental osmotic pressure of a $CaCl_2$ solution at room temperature.[2]
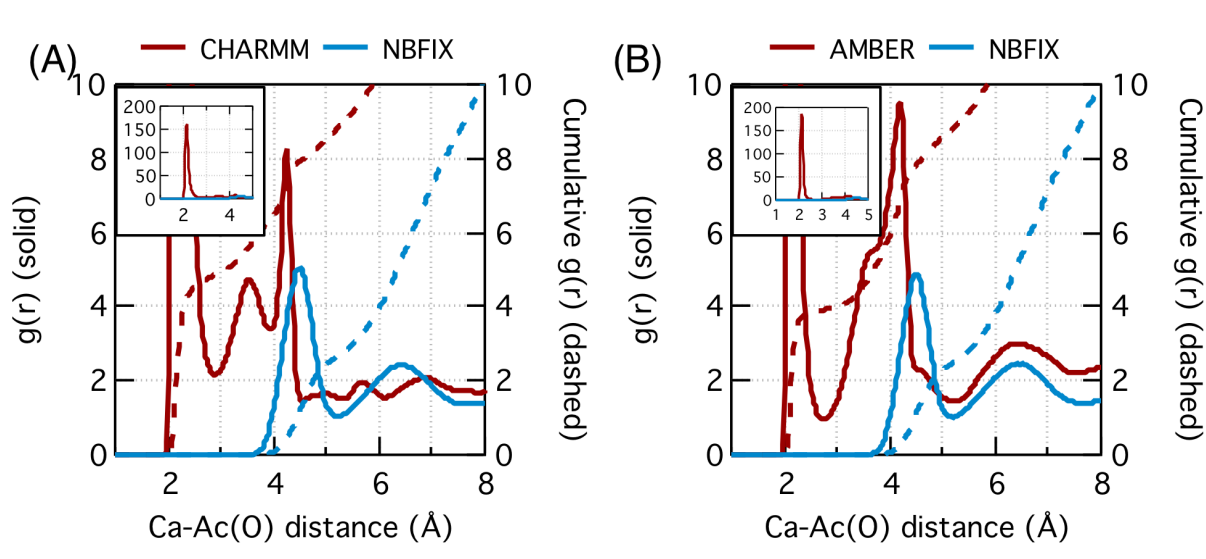
Figure S3: The effect of improved parameterization of the hydrated Ca^{2+} ion on radial distribution of acetate oxygen with respect to Ca^{2+} in simulations carried out using the CHARMM (A) and AMBER (B) force fields. (A) Radial distribution function (RDF) of acetate oxygen with respect to Ca^{2+} in MD simulation of a ∼2 m solution of $CaAc_2$. Blue and red lines indicate data obtained from the simulations of the same system performed using the CHARMM force field with and without our NBFIX corrections, respectively. Dashed lines show cumulative RDFs. The data were obtained by analyzing the last 50 ns of the respective production trajectories. (B) Same as in panel A but for the simulations carried out using the AMBER force field.
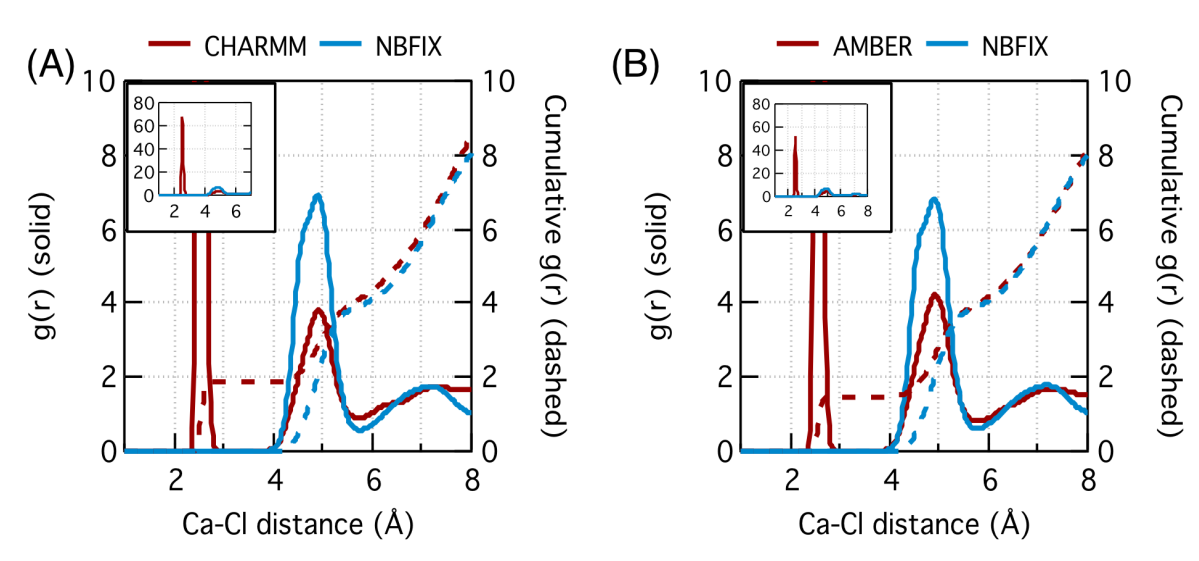
Figure S4: The effect of improved parameterization of the hydrated Ca^{2+} ion on radial distribution of Cl^- with respect to Ca^{2+} in simulations carried out using the CHARMM (A) and AMBER (B) force fields. (A) Radial distribution function (RDF) of Cl^- with respect to Ca^{2+} in MD simulation of a ~3 m solution of $CaCl_2$. Blue and red lines indicate data obtained from the simulations of the same system performed using the CHARMM force field with and without our NBFIX corrections, respectively. Dashed lines show cumulative RDFs. The data were obtained by analyzing the last 50 ns of the respective production trajectories. (B) Same as in panel A but for the simulations carried out using the AMBER force field.
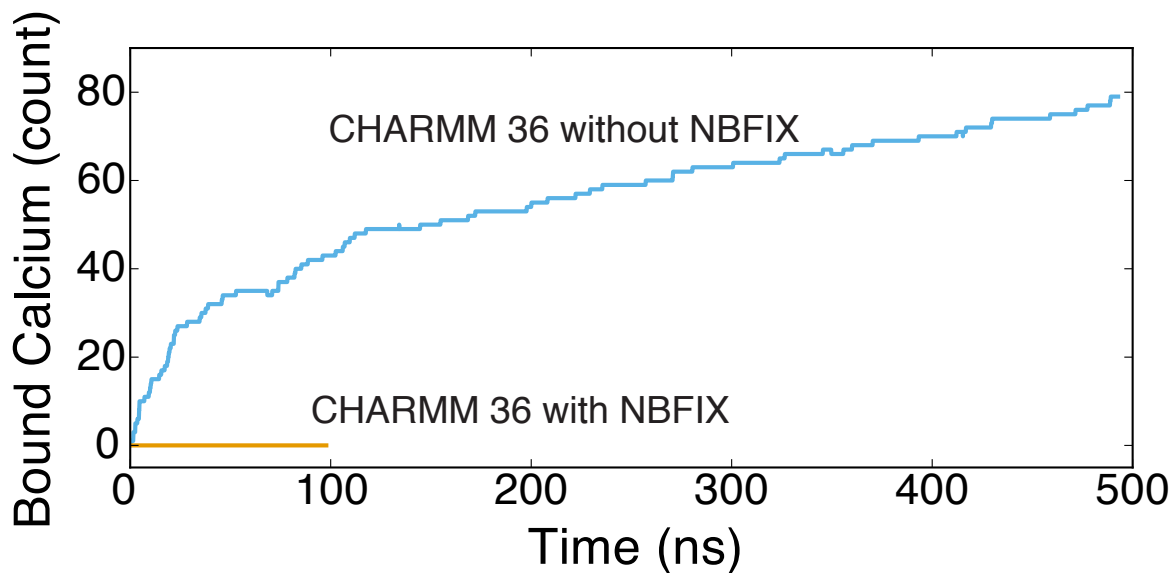
Figure S5: Binding of Ca^{2+} ions during the equilibration simulations of a 36-bp dsDNA fragment at 0.4 M KCl and 65 mM $CaCl_2$. The simulations were performed using either the combination of the CHARMM force field with our $Ca^{2+}(H_2O)_7$ model of hydrated Ca^{2+} ions (orange trace) or just the standard parameterization of the CHARMM force field (blue trace). A calcium ion was counted as bound to DNA if it was located within 0.3 nm of a phosphate oxygen atom of the DNA backbone. During the equilibration simulation employing the standard Ca^{2+} model, the bulk concentration of Ca^{2+} ions was maintained at 65 mM by adding additional Ca^{2+} and Cl^- ions to the system, compensating for permanent binding of Ca^{2+} to DNA. No calcium ions bind to DNA in the equilibration simulation performed using our improved model of calcium ions.

References

(1) Apelblat, A.; Korin, E. The vapour pressures of saturated aqueous solutions of magnesium, calcium, nickel and zinc acetates and molar enthalpies of solution of magnesium, calcium, zinc and lead acetates. *J. Chem. Thermodynamics* **2001**, *33*, 113–120.

(2) Robinson, R. A.; Stokes, R. H. *Electrolyte Solutions*; Butterworths scientific publications, 1959.