Supplementary Information (SI) for Soft Matter.

Supplementary Data and Figures for

Contraction response of a Polyelectrolyte Hydrogel to Nonuniformly Applied Electric Fields

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Simulation results of further work

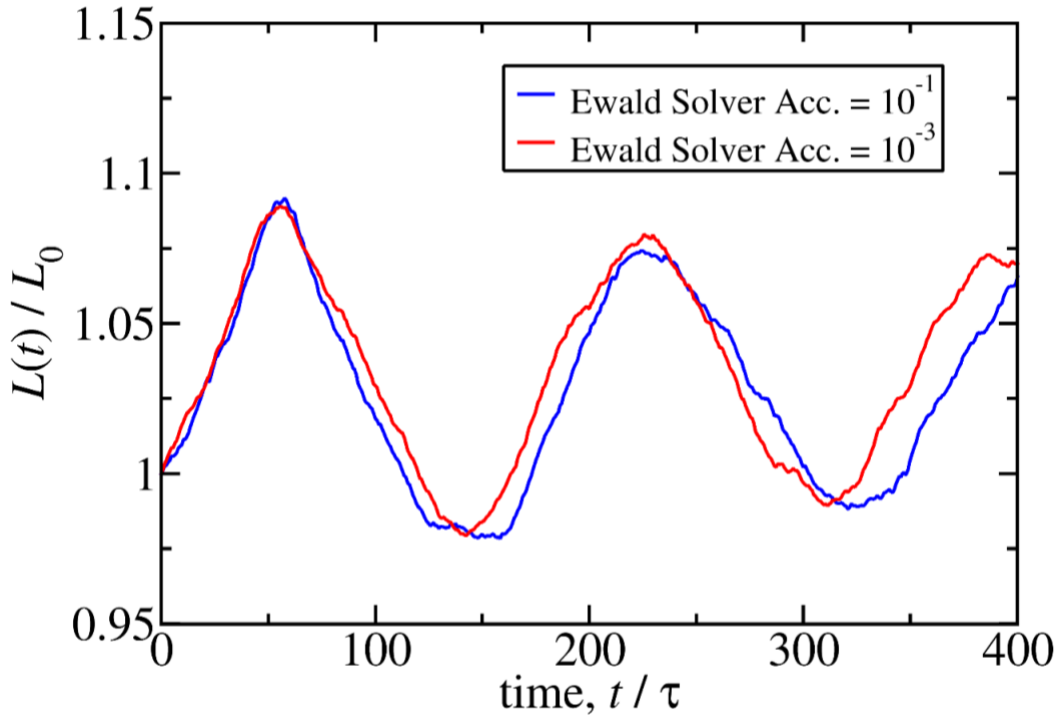

Figure 1: Electric field applied with amplitude of 0.5 and frequency of 1/100τ to hydrogel system constructed as 20, 5, 3 units of lattices repeated in the x̂ ŷ ẑ directions in implicit solvent simulations. With the same relaxation conditions, different Ewald solver accuracies considered (10^{-1} for blue, $10^{-3}$for red line) to validate if low Ewald solver accuracy changes hydrogels behavior qualitatively.

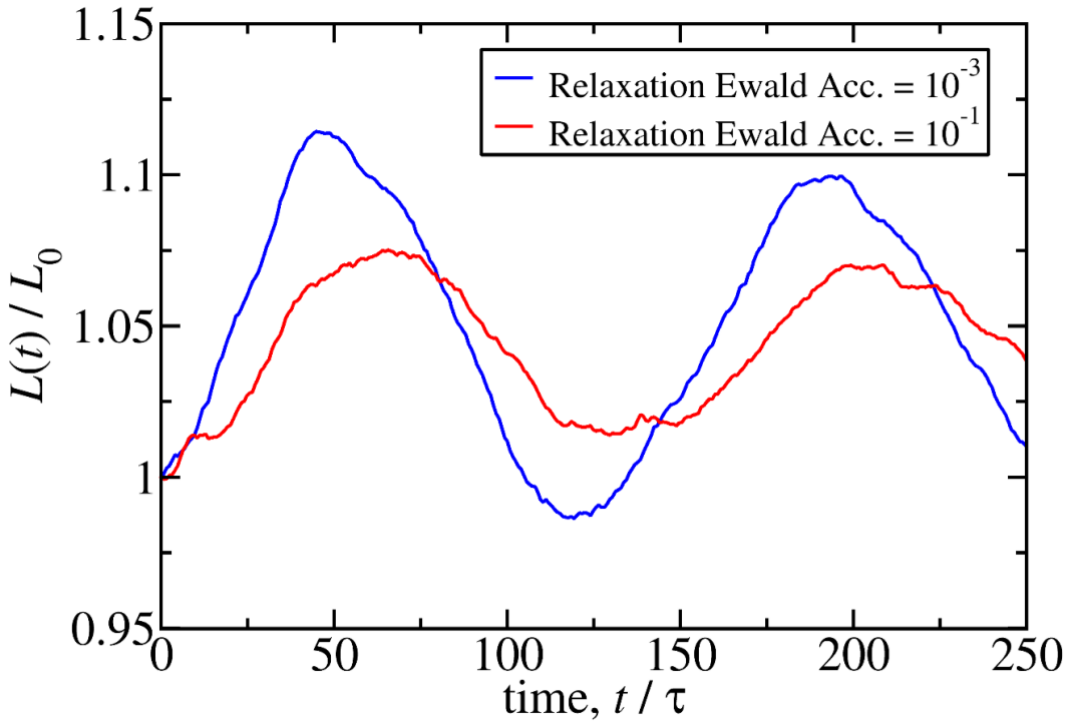

Figure 2: For hydrogel system constructed as 20, 5, 3 units of lattices repeated in the x̂ ŷ ẑ directions, different Ewald solver accuracies compared in relaxation stage for different reduced LJ temperature (T=1.0 [ε/k_b]) in implicit solvent simulations. The electric field is applied after relaxation with 0.5 amplitude and 1/100τ frequency. Qualitatively, there is no significant difference between different Ewald solver accuracies.

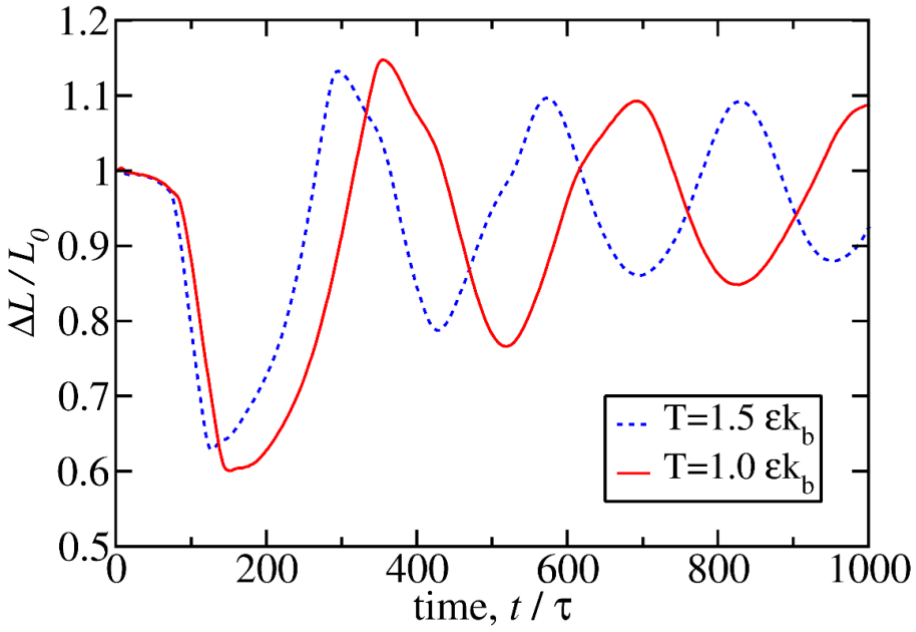

Figure 3: For the hydrogel system constructed as 40, 10, and 5 units of lattices repeated in the $\hat{x}$ $\hat{y}$ $\hat{z}$ directions, the effect of temperature was observed. The electric field was applied after relaxation with 4.0 amplitude and 1/100τ frequency. Qualitatively, there is no significant difference, and quantitatively, shrinkage increased by 2.81% (L_c 37.14% to 39.95%), and contraction time increased by 19% (τ_c 128 to 152 [t/τ]) when run at T=1.0 instead of T= 1.5 [ε/k_b].

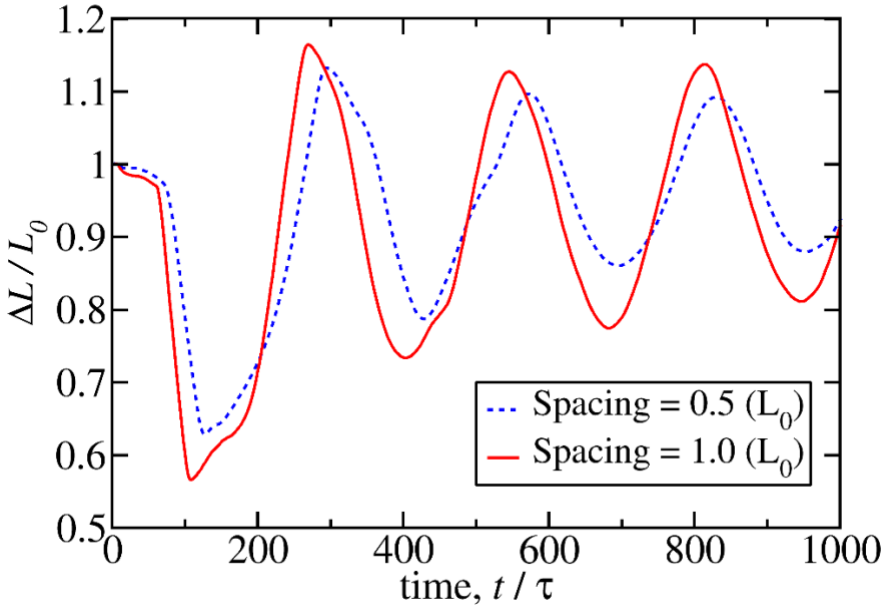

Figure 4: For the hydrogel system constructed as 40, 10, and 5 units of lattices repeated in the $\hat{x}$ $\hat{y}$ $\hat{z}$ directions in implicit solvent simulations, the effect of different spacing between two hydrogels was observed. An electric field is applied after relaxation with 4.0 amplitude and 1/100τ frequency. One with $0.5L_0$ and the other with $1.0L_0$ between two hydrogels to see if the electric field effect also impacts nearby hydrogels. Qualitatively, there is no significant difference, and because this is the case with no salt, it should also be ok for systems with salt because there is higher electrostatic screening. Quantitatively, shrinkage increased by 6.30% (L_c 37.14% to 43.44%), and contraction time decreased by 14% (τ_c 128 to 108 [t/τ]) when run at T=1.0 instead of T= 1.5 [ε/k_b].

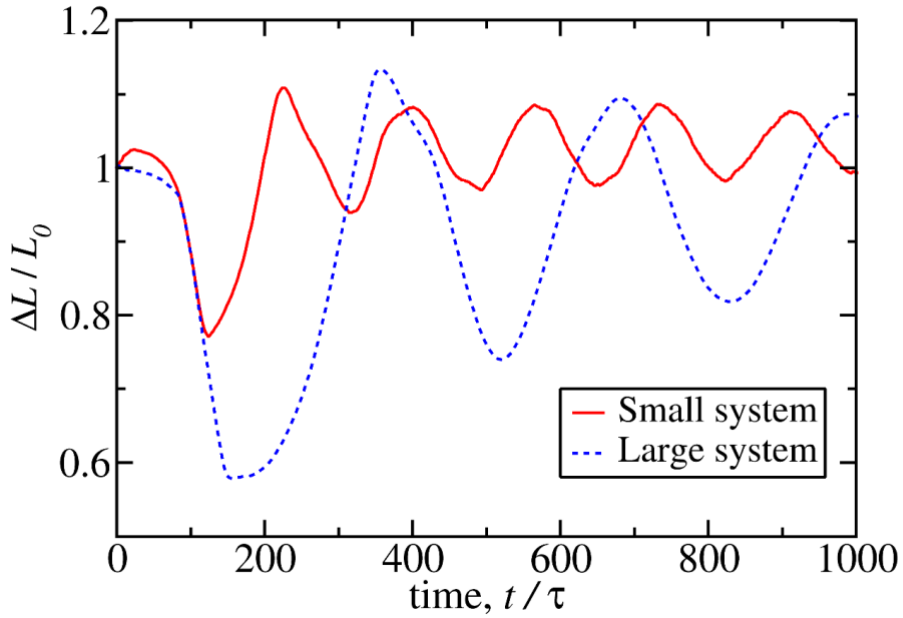

Figure 5: Comparison of small and large hydrogel systems, which are constructed as 40, 10, 5, and 20, 5, 3 units of lattices repeated in the $\hat{x}$ $\hat{y}$ $\hat{z}$ directions, respectively, in implicit solvent simulations. The electric field applied after relaxation with 4.0 amplitude and 1/100τ frequency where T=1.0 [ε/k_b]. Qualitatively, there is no significant difference, and quantitatively, shrinkage decreased by 20% (L_c 42.16% to 22.89%), and contraction time decreased by 21% (τ_c 156 to 123.2 [t/τ]) when a smaller system was investigated instead of a larger system.

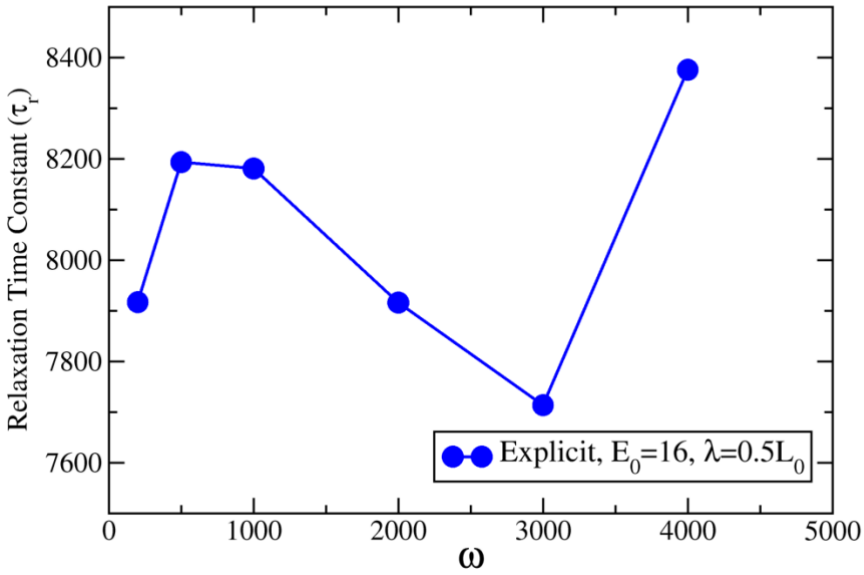

Figure 6: The retraction time of hydrogel in the slab in explicit solvent simulations. The relaxation time is obtained via an exponential fit in the form of f(x) = 1-Aexp (-t/B); here, A and B are fit parameters.

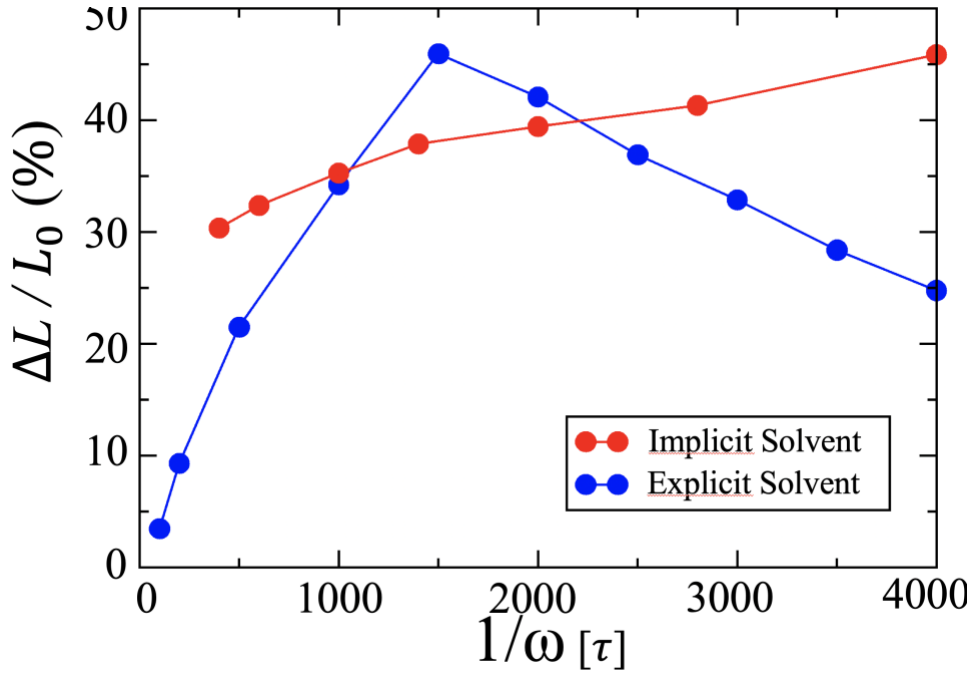

Figure 7: The contraction efficiency as a function of inverse frequency in explicit and implicit solvent simulations. E0 = 16 in reduced simulation units for all cases. However, the frequency axes for the implicit-simulation data are multiplied by 10 to provide a better visual presentation.

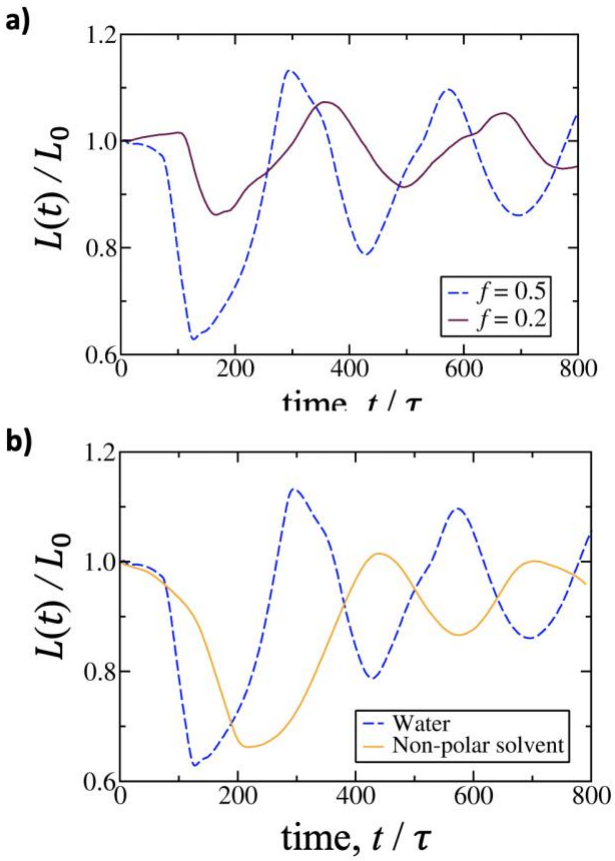

Figure 8: a) Contraction efficiency for two backbone charge fraction cases in implicit solvent simulations. The case with f=0.5 is used to obtain the data given in the main text. b) The contraction for two different dielectric cases. The case corresponding to water is used in the main text. In the non-polar case, the hydrogel is less swollen.

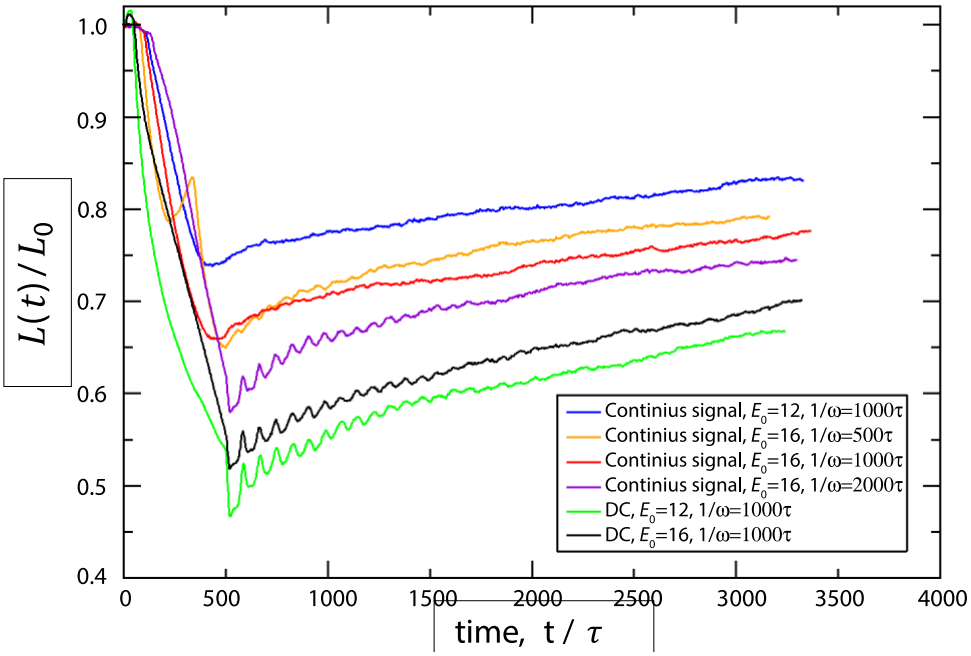

Figure 9: Contraction efficiency for various electric field signal schemes. In continuous signal, a complete sinusoidal signal is applied on the hydrogel slab in lambda-region (λ/L_0=0.5). For DC, a constant E-field is applied for the duration of the pulse. For all cases, f=0.5.

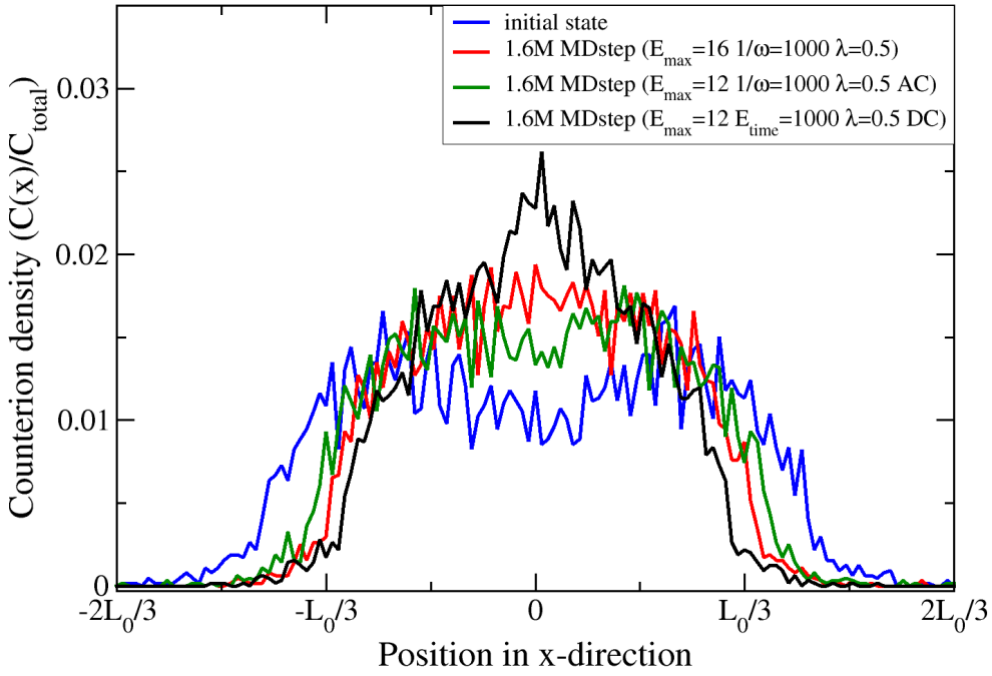

Figure 10: Normalized counterion density across the hydrogel slab at various time windows, corresponding to the initial (deformation-free) and field-exerted states.