Supporting Information for
Water Flux-Induced Reorientation of Liquid Crystals

Hadi Ramezani-Dakhel\textsuperscript{1,2}, Monirosadat Sadati\textsuperscript{1}, Rui Zhang\textsuperscript{1}, Mohammad Rahimi\textsuperscript{1}, Khia Kurtenbach\textsuperscript{1}, Benoît Roux\textsuperscript{2,*}, Juan J. de Pablo\textsuperscript{1,3,*}

\textsuperscript{1}Institute for Molecular Engineering, University of Chicago, Chicago, IL 60637, USA
\textsuperscript{2}Department of Biochemistry and Molecular Biology, University of Chicago, Chicago, IL 60637, USA
\textsuperscript{3}Argonne National Laboratory, Argonne, IL 60439, USA

*Correspondence to: B. Roux, E-mail: roux@uchicago.edu; J. J. de Pablo, E-mail: depablo@uchicago.edu

This PDF file includes:

- Materials and Methods
- Figures S1 to S12
- Table S1

Other supporting information for this manuscript includes the following:

- Movie S1
S1. Materials and Methods

S1.1. Standard Atomistic Molecular Dynamics Simulations. To elucidate the atomic-scale orientations of the molecules near the LC-aqueous interfaces and to uncover the distribution of the molecular entities, we performed atomistic molecular dynamics simulations. We followed our previously described procedure to setup the simulations, verify the convergence, and analyze the results.\textsuperscript{1} Briefly, we imposed a hybrid boundary condition on a thin film of 5CB in the nematic (25°C) and isotropic (40°C) phases by respectively contacting the upper and the lower planes of the film with an aqueous solution and vacuum. A periodic simulation box of dimensions 110×110×450 Å\textsuperscript{3} was prepared, with 8,000 molecules of 5CB and 24,000 pre-equilibrated liquid water molecules. The solutions of sodium chloride (NaCl) and sodium iodide (NaI) were created by placing monovalent ions (Na\textsuperscript{+}, Cl\textsuperscript{-}, and I\textsuperscript{-}) in random positions throughout the water phase, followed by removal of atomic overlaps through energy minimization or translational moves.

For 5CB, we rely on the united-atom force field of Tibero \textit{et al.}\textsuperscript{2} This force field is based on a classical Hamiltonian that was validated using experimental densities, orientational order parameter, and NMR residual dipolar couplings of 5CB.\textsuperscript{2} For ions, we rely on the classical force field of Joung and Cheatham, III.\textsuperscript{3} This force field was optimized against hydration free energies of solvated ions, lattice energies of crystals, and lattice constants of alkali halide salts crystals. For water, we use a thermodynamically consistent four-site model, namely TIP4Q, which was parameterized to reproduce the static dielectric constant, anomalous properties, and phase behavior of water with good overall agreement with experiments.\textsuperscript{4} Our earlier investigations of thermodynamic properties of the electrolyte solutions including surface adsorption energy, solvation free energy, and surface tension shows that the force fields parameters of TIP4Q water
and monovalent ions used herein are fully compatible without any further adjustments of the parameters.\(^5\)

The standard simulation protocol involved unrestrained molecular dynamics simulations using the Nanoscale Molecular Dynamics package (NAMD)\(^6\) in the canonical ensemble (NVT). We used a time step of 2 fs, and the Langevin thermostat with a damping coefficient of 1 ps\(^1\). A spherical cutoff of van der Waals interactions at 12 Å with a switching distance at 10 Å, and summation of Coulomb interactions using the Particle Mesh Ewald (PME) method with a high accuracy of 10\(^{-6}\) kcal/mol were employed throughout the equilibration and production runs. All system were subjected to multiple intervals of 50 ns MD simulations (6×50 ns for the nematic phase and 4×50 ns for the isotropic phase), accompanied by on-the-fly analysis of molecular organization and thermodynamic data.\(^1\)

**S1.2. Umbrella Sampling Potential of Mean Force Calculations.** To perform the umbrella sampling potential of mean force (PMF) calculations related to the relocation of a water molecule from the pure water phase and 2 M solutions of NaI and NaCl to the vacuum phase, we used pre-equilibrated slabs of water (a layer of water in contact with vacuum on the upper and lower planes), NaI, and NaCl solutions with 10,000 water molecules and an approximate box dimensions of 67×67×167 Å\(^3\). PMF calculations of a water molecule that leaves the water phase and resides inside the bulk LC phase involved an equilibrated system of 8,000 5CB molecules, and 24,000 water molecules (see section S1.1).

For the aqueous solution-vacuum calculations, we applied a harmonic biasing potential with a force constant of 5 kcal/mol·Å\(^2\) to the z-distance between the center-of-mass of the water phase (10,000 water molecules) and the oxygen atom of a single water molecule. The reaction coordinate were varied successively from \(z = -20\) to \(z = 20\) Å every 1 Å resulting in a total of 41
windows. The water-vacuum interface is approximately located at \(z = 0\), thereby \(z = -20\ \text{Å}\) corresponds to a water molecule located in the bulk region and \(z = 20\ \text{Å}\) indicates an isolated water molecule in vacuum. In total, we generated 123 independent simulations (41 windows for three systems including pure water, NaI solution, and NaCl solution), and subjected each individual window to a 3 ns equilibration followed by a 7 ns production molecular dynamics simulations. The structure and energy of the system immediately converged within an initial period of < 3 ns due to the absence of chain molecules.

For the water-LC system, we applied a harmonic biasing potential with a force constant of 5 kcal/mol·Å\(^2\) to the \(z\)-coordinate of the oxygen atom of a water molecule. The center of the harmonic potential were varied sequentially from \(z = -40\) to \(z = 20\ \text{Å}\) every 1 Å with a total of 61 windows. The water-LC interface is approximately positioned around \(z = 0\). Accordingly, \(z = -40\ \text{Å}\) corresponds to a water molecule that fully resides inside the LC phase and \(z = 20\ \text{Å}\) corresponds to the bulk water phase.

The protocol of the free energy calculations involved simulations in the canonical ensemble (NVT) with a time step of 2 fs. The Langevin thermostat with the damping coefficient of 1 ps\(^{-1}\) was engaged to control the temperature. The pair-wise van der Waals interactions were spherically cutoff at 12 Å, the summation of Coulomb interactions were handled using the Particle Mesh Ewald (PME) method with a high accuracy of \(10^{-6}\) kcal/mol. The trajectories of the collective variables were buffered and synchronize every 100 steps. The umbrella sampling simulations were unbiased together using weighted-histogram-analysis-method (WHAM)\(^{7,8}\) to obtain the full PMF profiles (Figure 2). We used the program WHAM\(^9\) with energy tolerance of \(10^{-12}\) accompanied with self-written scripts to perform the unbiasing calculations.
S1.3. Continuum-Level Simulations of the Time-Dependent Density Profiles and Water Flux According to Smoluchowski Equation. To elucidate the time-dependent evolution of the profile of water density inside the LC cell after addition of the electrolytes, and to compute the flux of water through the LC-aqueous interface, we solved the Smoluchowski (drift-diffusion) equation:\textsuperscript{10}

\[
\frac{\partial \rho}{\partial t} = \nabla \cdot (D \nabla \rho) - \nabla \cdot (\zeta^{-1} \vec{F} \rho) \tag{S1}
\]

here, \( \rho \) is the water density, \( t \) is time, \( D \) is the diffusion constant, \( \zeta^{-1} = D/k_B T \) is the mobility, \( \vec{F} = -\nabla W \) is the mean force, and \( W \) is the potential of the mean force (PMF). By assuming a location-independent diffusion constant \( D \), the Smoluchowski equation of a one-dimensional system, i.e. the thin LC film, can be reformulated in the form:

\[
\frac{\partial \rho}{\partial t} = D \frac{\partial^2 \rho}{\partial z^2} + \frac{D}{k_B T} \frac{dW}{dz} \frac{\partial \rho}{\partial z} + \frac{D}{k_B T} \frac{d^2W}{dz^2} \rho \tag{S2}
\]

here, \( z \) is the distance from the interface (we used \( z \) instead of \( D \) to avoid confusions with the diffusion constant). We approximated the solution of the Smoluchowski equation numerically using a finite difference approach, and used our in-house codes to perform the calculations.

First, we resolved the equilibrium profile of the water density inside a LC film in contact with pure water at the interface. In this simulation, the potential of the mean force corresponding to the relocation of a water molecule from the bulk water to the bulk liquid crystal phase, i.e. the \( W \) term in Smoluchowski equation, was obtained using the umbrella sampling molecular dynamics simulations (see section S1.2 and Figure 2A). Because the profile of the PMF follows a \( \tanh \)-like behavior, we fitted a \( \tanh \) function in the form \( W = \frac{A}{2} \tanh(B(z - z_0)) + \frac{A}{2} \) to the computed PMF curves, and employed that profile and its derivatives in our calculations. In this function, \( A \) is the free energy necessary for relocation of a single water molecule from the pure
water phase to the LC phase (Figure 2), \( B = 0.5 \), and \( z_0 = 5 \). Next, we simulated the time-dependent evolution of the density profile and water flux induced by addition of salts to the aqueous phase. Accordingly, the equilibrium profile of the water density obtained in the previous step served as initial configuration for the simulation of water flux and density profile. Immediately after addition of the salts, the water molecules began to diffuse through the LC-aqueous interface resulting in a significant instantaneous flux of water (Movie S1).

**S1.4. Estimation of the Equilibrium Time of the LC-Aqueous Interface after Addition of NaI Electrolyte.** To estimate the time necessary to reach the equilibrium concentration of water inside the LC film after addition of NaI electrolyte, we continued our continuum simulations of water diffusion for \( \sim 5 \times 10^4 \) \( \mu s \) (Figure S12). Within the \( 5 \times 10^4 \) \( \mu s \) of immersion under salt solution, the water flux reaches a quasi-steady state limit with very small variations. Accordingly, we assumed that the water flux would remain unchanged after \( 5 \times 10^4 \) \( \mu s \), and computed the time required for the net flux of water through the interface to reach zero using the following expression:

\[
t = 5 \times 10^4 + \frac{N_{\text{wat}}^{t=0}(1-a_{\text{wat}}^{\text{NaI}})-N_{\text{av}}A\int_{5 \times 10^4}^{t} f(t)dt}{N_{\text{av}}f(t>5 \times 10^4)A} \tag{S3}
\]

here, \( t \) is time in microseconds, \( N_{\text{wat}}^{t=0}(1-a_{\text{wat}}^{\text{NaI}}) \) denotes the target number of water molecules that would eventually leave the LC phase, \( N_{\text{wat}}^{t=0} \) is the initial total number of water molecules in a LC grid in experiments, \( a_{\text{wat}}^{\text{NaI}} \) is the water activity of the NaI solution, \( N_{\text{av}} \) is the Avogadro’s number, \( A \) is the cross-sectional area of a LC grid, \( f(t) \) is the time-dependent water flux, \( f(t > 5 \times 10^4) \) is the magnitude of the water flux at \( t = 5 \times 10^4 \) \( \mu s \). Accordingly, we estimated an equilibration time of \( \sim 3.8 \) hours for the LC-aqueous interface.
S1.5. Continuum Simulations of Flux-Induced Anchoring Transitions According to Beris-Edwards Equation. To explain the evolution of the director field of a hybrid LC channel under an instantaneous flux of water, we solved the Berris-Edwards hydrodynamic equation:

\[(\partial_t + \mathbf{u} \cdot \nabla) \mathbf{Q} - \mathbf{R}(\mathbf{W}, \mathbf{Q}) = \Gamma \mathbf{H}\]  \hspace{1cm} (S4)

here, \(\mathbf{u}\) is the velocity field, \(\mathbf{Q}\) is the tensorial order parameter, \(\mathbf{W}\) is the gradient of the velocity \((W_{ij} = \partial_j u_i)\), \(\Gamma\) is a constant related to rotational viscosity \(\gamma_1\) \((\Gamma = 2S^2/\gamma_1\), where \(S\) is the scalar order parameter of the nematic LC phase equivalent to the largest eigenvalue of the \(\mathbf{Q}\) tensor), \(\mathbf{H}\) is the molecular field related to the free energy of the system, and \(\mathbf{R}(\mathbf{W}, \mathbf{Q})\) is the advection term.

The advection term \(\mathbf{R}(\mathbf{W}, \mathbf{Q})\) is given by:

\[
\mathbf{R}(\mathbf{W}, \mathbf{Q}) = (\xi \mathbf{A} + \mathbf{\Omega})(\mathbf{Q} + \mathbf{I}/3) + (\mathbf{Q} + \mathbf{I}/3)(\xi \mathbf{A} - \mathbf{\Omega}) - 2\xi(\mathbf{Q} + \mathbf{I}/3)\text{Tr}(\mathbf{WQ})
\]  \hspace{1cm} (S5)

where, \(\xi\) is a material constant related to the molecular aspect ratio, \(\mathbf{I}\) is the identity matrix, \(\mathbf{A}\) is the strain rate tensor \(A_{ij} = (\partial_i n_j + \partial_j n_i)/2\), and \(\mathbf{\Omega}\) is the vorticity tensor \(\Omega_{ij} = (\partial_i n_j - \partial_j n_i)/2\).

The molecular field \(\mathbf{H}\) drives the system towards a thermodynamic equilibrium with rate \(\Gamma\),\(^{11}\) and is defined as:

\[
H_{ij} = -\frac{1}{2} \left[ \frac{\delta f}{\delta Q_{ij}} + \left( \frac{\delta f}{\delta Q_{ij}} \right)^T \right] + \frac{\delta_{ij}}{3} \text{Tr} \left( \frac{\delta f}{\delta Q_{ij}} \right)
\]  \hspace{1cm} (S6)

where \(f = f_{\text{LandG}} + f_{\text{el}}\) is the free energy density of the system. \(f_{\text{LandG}}\) corresponds to the Landau-de Gennes free energy which describes the short-range interactions, and \(f_{\text{el}}\) determines the long-range elastic energy. These two free energy terms read:

\[
f_{\text{LandG}} = \frac{A_0}{2} \left( 1 - \frac{\nu}{3} \right) Q_{ij} Q_{ij} - \frac{A_0 U}{3} Q_{ij} Q_{jk} Q_{kl} + \frac{A_0 U}{4} \left( Q_{ij} Q_{ij} \right)^2
\]  \hspace{1cm} (S7)

\[
f_{\text{el}} = \frac{1}{2} L \partial_k Q_{ij} \partial_k Q_{ij}
\]  \hspace{1cm} (S8)

where \(A_0\) and \(U\) are constants, \(L\) is the molecular field related to the free energy of the system.
here, $A_0$ and $U$ are the thermodynamic parameters that define the equilibrium scalar order parameter in an undistorted LC bulk. $L$ is the elastic constant under a single-constant approximation.

To solve the Beris-Edwards equation for a LC film, one needs to resolve the velocity field of the LC phase induced by the flow of water due to addition of salts to the aqueous phase. For that purpose, we assumed an exclusive momentum transfer from the diffusing water molecules to the 5CB molecules, and obtained the corresponding velocity field of the LC phase (Figure 4A). The velocity field $u$ in a given position inside the LC film is computed according to $u = \frac{J \cdot m_{Wat}}{\rho_{LC}}$, where $J$ is the water flux, $m_{Wat}$ is the molar mass of water, and $\rho_{LC}$ is the mass density of 5CB. The initial profile of water flux $J$ inside the LC film is obtained using the Smoluchowski equation (see section S1.3). Because the profile of water flux (and water velocity) follows an exponential behavior (Figure 4A), we fitted an exponential function $u = Ce^{-D}$ to the computed velocity profile, and employed that profile in our calculations. Here, $C$ is a constant and $D$ is the distance from the interface.

To solve the Beris Edwards equation, we adopted our previously reported finite difference method and used our in-house codes. The initial configuration of the LC film was set to a typical splay-bend structure. The system was then allowed to evolve until a steady state configuration was reached. The elastic energy was then calculated as a function of the magnitude of the maximum water flux at the LC-aqueous interface (Figure 4B).
**Figure S1.** Orientational features of a nematic nanometer-sized 5CB thin film with the vacuum and sodium iodide (NaI) solution boundary conditions, according to molecular dynamics simulations. (A) Color-coded profile of second Legendre polynomial $P_2(z) = (2 \cos^2 \theta - \frac{1}{2})$. (B) Profile of scalar order parameter $S$ obtained from diagonalization of $Q$ tensor. (C) Components of the nematic director $n_z$ and $n_{xy} = \sqrt{(n_x^2 + n_y^2)}$ at a given distance from the vacuum and aqueous interface. We observed a splay-bend geometry with continuous transition of the nematic director from a perpendicular orientation at the LC-vapor interface to a parallel orientation near the aqueous interface.
Figure S2. Orientational features of a nematic 5CB film restrained between vacuum and aqueous electrolyte of sodium chloride (NaCl) obtained from molecular dynamics simulations. (A) Color-coded profile of second Legendre polynomial $P_2(z) = \frac{3}{2} \cos^2 \theta - \frac{1}{2}$ . (B) Profile of scalar order parameter $S$ obtained from diagonalization of $Q$ tensor. (C) Components of the nematic director $n_z$ and $n_{xy} = \sqrt{n_x^2 + n_y^2}$ at a given distance from vacuum and aqueous interfaces. The error-bars here correspond to the standard deviations of the computed values over the final 30 ns of MD simulations.¹
Figure S3. Ensemble-average profiles of the second-order Legendre polynomial $\langle P_2(z) \rangle$ order parameter of nematic and isotropic thin films of 5CB computed with respect to the surface normal $\hat{z} = (0,0,1)$. (A) vacuum and 2M solution of sodium chloride, and (B) vacuum and 2M ionic solution of sodium iodide boundary conditions.
**Figure S4.** Two-dimensional probability distribution functions of the molecular orientation $P(z, \cos \beta)$ of a 5CB thin film walled by vacuum and sodium iodide aqueous electrolyte at the bottom and top boundaries. Here $z$ represents the vertical distance from LC-aqueous interface, and $\beta$ denotes the angle between the long molecular axis of 5CB and $\hat{z} = (0,0,1)$. (A) The density of molecular orientations in at 25°C (nematic phase of 5CB). (B) Contour maps of probability distribution functions at 40°C (isotropic phase). The probability of orientational distribution function shows an anisotropic distribution around $\cos \beta = 0$ with a slight shift towards positive values of $\cos \beta$. Near the free interface, the orientational distribution function is characterized by two distinct antiparallel molecular double-layer.
Figure S5. Two-dimensional histograms of molecular orientation $P(z, \cos \beta)$ of a 5CB thin film bound by vacuum and a 2 M solution of sodium chloride (NaCl). Here $z$ represents the vertical distance from LC-aqueous interface, and $\beta$ denotes the angle between the long molecular axis of 5CB and $\hat{z} = (0,0,1)$. (A) The density of molecular orientations at 25°C (nematic phase of 5CB). (B) Contour maps of probability distribution functions at 40°C (isotropic phase).
Figure S6. Molecular organizations of an isotropic thin film of 5CB bound with vacuum on the lower plain and the 2 M solution of sodium iodide on the upper plane, obtained from atomistic molecular dynamics simulations. (A) Color-coded profile of second Legendre polynomial $\langle P_2(z) \rangle = \left( \frac{3}{2} \cos^2 \theta - \frac{1}{2} \right)$ where $\theta$ in the angle between long molecular axis of 5CB and $z = (0,0,1)$ (B) Profile of scalar order parameter $S$ obtained by construction and diagonalization of $Q$ tensor. (C) Profile of individual components of the nematic director $n_z$ and $n_{xy} = \sqrt{(n_x^2 + n_y^2)}$ of isotropic hybrid channel of 5CB. The error-bars show the standard deviations of the calculated values over the final 30 ns block of the MD simulations.
Figure S7. Molecular organizations of an isotropic thin film of 5CB exposed to vacuum on the bottom and 2M solution of sodium chloride on top, established by molecular dynamics simulations. (A) Color-coded profile of second Legendre polynomial $\langle P_2(z) \rangle = \langle \frac{3}{2} \cos^2 \theta - \frac{1}{2} \rangle$ where $\theta$ is the angle between long molecular axis of 5CB and $\hat{z} = (0,0,1)$ (B) Profile of scalar order parameter $S$ obtained by construction and diagonalization of $Q$ tensor. (C) Profile of individual components of nematic director $n_z$ and $n_{xy} = \sqrt{(n_x^2 + n_y^2)}$ of the thin film with homeotropic anchoring at the LC-vacuum interface and planar alignment near the aqueous interface.
Figure S8. The normalized probability of molecular distribution, and the average polarization density of the 5CB and water near the LC-aqueous and aqueous-vacuum interfaces. (A) Normalized probability of finding selected atoms of the main functional groups including nitrogen atom of the nitrile group, oxygen of water molecule, sodium ion, and iodide ion. (B) Polarization density profiles of water molecules in a 2 M solution of sodium iodide (NaI) in contact with 5CB on one side and vacuum on the other end, at 25 and 40°C. Specific interfacial arrangements of the hydrated cations and anions results in an electric field that perturbs the original orientation of the interfacial water molecules. The generated internal electric field exerts an orienting torque on molecular dipole of water molecules which in turn augments the average polarization density of water phase near the interfaces. This amplification of the polarization density of water highly depends on specific localization of oppositely charged ionic species in
aqueous phase. Upon solvation, sodium iodide dissociate into distinct layers of anion-rich and cation-rich domains near the interfaces to create an electric field which is strong enough to reorient water molecules but not strong enough to permanently modify interfacial alignment of the LC phase.
**Figure S9.** Distribution profiles and average polarization density of molecules and ions obtained from 300 ns molecular dynamics simulations. (A) Relative probability of finding 5CB, water, sodium, and chloride near interfacial regions of the LC-aqueous and aqueous-vacuum. (B) Average polarization density of interfacial water molecules and nematic (25°C) and isotropic (40°C) phases of 5CB in the presence of the sodium chloride (NaCl) solution at equilibrium. In contrast to sodium iodide, the solvated sodium chloride forms less distinct ionic-rich layers and thereby the generated internal electric field marginally modifies the polarization density of water molecules near the interfaces.
Figure S10. Anchoring strength profiles $W^A_2(z)$ of 5CB at vacuum, water, sodium chloride solution, and sodium iodide solutions. (A) nematic and (B) isotropic phases. Substitution of the distilled water with the ionic solutions of sodium iodide and sodium chloride slightly lowers the anchoring strength of the nematic LC at the aqueous phase. In isotropic phase, however, the anchoring coefficient profiles of 5CB at the pure water and ionic solutions remain essentially identical.
Figure S11. Potential of mean force (PMF) for the transfer of a chloride ion from the bulk water to the bulk LC phase. Ions transferring to the nonpolar phase tend to remain surrounded by a first solvation shell of water at all times due to the prohibitively large hydration free energy. The free energy cost for translocation of such a partially hydrated chloride ion into the LC phase is approximately 12 kcal/mol, which corresponds to nanomolar concentrations of ions in the LC phase.
Figure S12. The time-dependent flux of water through the LC-aqueous interface after immersion under 2 M solution of NaI aqueous electrolytes. Within 50,000 µs of immersion under salt solution, the water flux reaches a quasi-steady state limit with very small variations. Accordingly, we assumed that the water flux would remain unchanged after $5 \times 10^4$ µs, and computed the time required for the net flux of water through the interface to reach zero (see section S1.4)
**Table S1.** The intimate relationship between the water activity of simple solutions, the magnitude of the instantaneous water flux through the LC-water interface, and the reorientation of liquid crystals allows design of new solutes with triggerable properties. Here, the activity data of NaI, LiCl, NaCl, and NaOH solutions are taken from reference,\textsuperscript{13} and the activity data of NaClO\textsubscript{4} solution is taken from reference.\textsuperscript{14} The triggerable properties of NaClO\textsubscript{4}, and NaOH solutions have been demonstrated in previous studies.\textsuperscript{15,16}

| Solute    | Water activity of near saturated solutions | Triggers anchoring transition (below 2 M concentration) |
|-----------|-------------------------------------------|--------------------------------------------------------|
| NaI       | 0.382                                     | Yes                                                   |
| LiCl      | 0.113                                     | Yes                                                   |
| NaClO\textsubscript{4} | 0.245                          | Yes                                                   |
| NaCl      | 0.753                                     | No                                                    |
| NaOH      | 0.082                                     | Yes                                                   |
**Movie S1.** The movie shows (left) time-dependent profile of water density inside the liquid crystal film after addition of NaCl (green) and NaI (blue) salts (up to 1 ms). The red line shows the equilibrium profile of water density prior to addition of salts. (right) time-dependent flux of water through the LC-aqueous interface after addition of 2 M concentration of NaI (navy) and NaCl (green) electrolytes.

**Supporting References**

(1) Ramezani-Dakhel, H.; Sadati, M.; Rahimi, M.; Ramirez-Hernandez, A.; Roux, B.; de Pablo, J. J. Understanding Atomic-Scale Behavior of Liquid Crystals at Aqueous Interfaces. *J. Chem. Theory Comput.* **2017**, *13*(1), 237.

(2) Tiberio, G.; Muccioli, L.; Berardi, R.; Zannoni, C. Towards in Silico Liquid Crystals. Realistic Transition Temperatures and Physical Properties for n-Cyanobiphenyls via Molecular Dynamics Simulations. *ChemPhysChem* **2009**, *10*(1), 125.

(3) Joung, I. S.; Cheatham, T. E. Determination of alkali and halide monovalent ion parameters for use in explicitly solvated biomolecular simulations. *J. Phys. Chem. B* **2008**, *112*(30), 9020.

(4) Alejandre, J.; Chapela, G. A.; Saint-Martin, H.; Mendoza, N. A non-polarizable model of water that yields the dielectric constant and the density anomalies of the liquid: TIP4Q. *Phys. Chem. Chem. Phys.* **2011**, *13*(44), 19728.

(5) Whitmer, J. K.; Joshi, A. A.; Carlton, R. J.; Abbott, N. L.; de Pablo, J. J. Surface Adsorption in Nonpolarizable Atomic Models. *J. Chem. Theory Comput.* **2014**, *10*(12), 5616.

(6) Phillips, J. C.; Braun, R.; Wang, W.; Gumbart, J.; Tajkhorshid, E.; Villa, E.; Chipot, C.; Skeel, R. D.; Kale, L.; Schulten, K. Scalable molecular dynamics with NAMD. *J. Comput. Chem.* **2005**, *26*(16), 1781.
(7) Kumar, S.; Rosenberg, J. M.; Bouzida, D.; Swendsen, R. H.; Kollman, P. A. Multidimensional Free-Energy Calculations Using the Weighted Histogram Analysis Method. *J. Comput. Chem.* **1995**, *16* (11), 1339.

(8) Roux, B. The Calculation of the Potential of Mean Force Using Computer-Simulations. *Comput. Phys. Commun.* **1995**, *91* (1-3), 275.

(9) Grossfield, A. *WHAM: an implementation of the weighted histogram analysis method, version 2.0.9*, 2013.

(10) Smoluchowski, M. V. Über Brownsche Molekularbewegung unter Einwirkung äußerer Kräfte und den Zusammenhang mit der verallgemeinerten Diffusionsgleichung. *Ann. d. Physik* **1915**, *48*, 1103.

(11) Denniston, C.; Orlandini, E.; Yeomans, J. M. Lattice Boltzmann simulations of liquid crystal hydrodynamics. *Phys Rev E* **2001**, *63* (5), 056702.

(12) Zhang, R.; Roberts, T.; Aranson, I. S.; de Pablo, J. J. Lattice Boltzmann simulation of asymmetric flow in nematic liquid crystals with finite anchoring. *J. Chem. Phys.* **2016**, *144* (8), 084905.

(13) Barbosa-Canovas, G. V.; Fontana, A. J.; Schmidt, S. J. In *Institute of Food Technologists Series*; John Wiley & Sons: Hoboken, 2007.

(14) Toner, J. D.; Catling, D. C. Water activities of NaClO4, Ca(ClO4)(2), and Mg(ClO4)(2) brines from experimental heat capacities: Water activity > 0.6 below 200 K. *Geochim. Cosmochim. Acta* **2016**, *181*, 164.

(15) Carlton, R. J.; Gupta, J. K.; Swift, C. L.; Abbott, N. L. Influence of Simple Electrolytes on the Orientational Ordering of Thermotropic Liquid Crystals at Aqueous Interfaces. *Langmuir* **2012**, *28* (1), 31.
(16) Carlton, R. J.; Ma, C. D.; Gupta, J. K.; Abbott, N. L. Influence of Specific Anions on the Orientational Ordering of Thermotropic Liquid Crystals at Aqueous Interfaces. *Langmuir* **2012**, 28 (35), 12796.