

## Supplementary Information

### Ising Model

The water dipoles in the water chain were treated as lattice sites with two orientations ( $\sigma = +1$  or  $-1$ ) and the short-range interactions between them were included. The energy of the water dipole chain is given by the Hamiltonian Function  $\mathcal{H} = -\sum_{\langle i,j \rangle} J_{ij} \sigma_i \sigma_j - H \sum_{i=1}^N \sigma_i$ , where  $J_{ij}$  is the interaction of any two adjacent water dipoles  $i$  and  $j$ ,  $H$  is the external effect on each water dipole, namely the electrostatic interaction between the imposed charge and water dipoles,  $N$  is the number of water dipoles in the water chain, and the water molecule nearest to the charge was set as the first dipole ( $i = 1$ ). Assuming that the imposed charge only has an interaction of  $F$  with the first dipole and using the periodic boundary condition ( $\sigma_{N+1} = \sigma_1$ ), the Hamiltonian Function can be written as  $\mathcal{H} = -\sum_{i=1}^N J_{i,i+1} \sigma_i \sigma_{i+1} - F$ , and the steady state of the water dipole chain is at the minimum of  $\mathcal{H}$ . Considering that  $\sigma_i \in \{+1, -1\}$ ,  $\mathcal{H}$  gets the minimum when all the water dipoles have the same orientation without external charge. If a charge was imposed near the first dipole in the ordered dipole chain, the first dipole would change its orientation and finally all the dipoles would reverse their orientation to get the minimum of  $\mathcal{H}$ .

### Euler-Lagrange Equation

The Lagrangian of a chain containing  $N$  ( $N \rightarrow \infty$ ) water dipoles with an imposed charge  $q$  can be written as

$$L = \sum_{i=1}^N \frac{I}{2} \dot{\phi}_i^2 - \left( \sum_{i=1}^N \left( -\frac{1}{4\pi\epsilon_0 r^3} (\vec{\mu}_{i-1} \cdot \vec{\mu}_i + \vec{\mu}_i \cdot \vec{\mu}_{i+1}) \right) + \sum_{i=1}^N U_i \right) \quad (1)$$

A negative charge  $-q$  was assigned far away to neutralize the system. The imposed external dipole across the system, formed by  $q$  and  $-q$  ( $q = e_0 e, e_0 > 0$ ), can be split into  $N$  dipoles assigned nearby each water molecule. Each divided dipole is formed by two opposite charges  $q$  and  $-q$  with a distance of  $r$ . As a result,  $U_i = -\frac{6.83e_0\mu^2}{2\pi\epsilon_0 r^3} \cos \phi_i$ . In this case, eqn (1) becomes

$$L = \sum_{i=1}^N \frac{I}{2} \dot{\phi}_i^2 + \frac{\mu^2}{4\pi\epsilon_0 r^3} \sum_{i=1}^N [\cos(\phi_i - \phi_{i-1}) + \cos(\phi_{i+1} - \phi_i)] + \frac{6.83e_0\mu^2}{2\pi\epsilon_0 r^3} \sum_{i=1}^N \cos \phi_i \quad (2)$$

The Euler-Lagrange Equation  $\frac{d}{dt} \left( \frac{\partial L}{\partial \dot{\phi}_i} \right) = \frac{\partial L}{\partial \phi_i}$  gives

$$I \ddot{\phi}_i + \frac{\mu^2}{4\pi\epsilon_0 r^3} (2\phi_i - \phi_{i-1} - \phi_{i+1}) + \frac{6.83e_0\mu^2}{2\pi\epsilon_0 r^3} \sin \phi_i = 0 \quad (3)$$

For the continuum model,  $\phi_i(t) = \phi(ir, t) = \phi(x, t)$ , using Taylor expanding around  $x$ , we have

$$2\phi(x, t) - \phi(x - r, t) - \phi(x + r, t) = -r^2 \frac{\partial^2 \phi(x, t)}{\partial x^2} \quad (4)$$

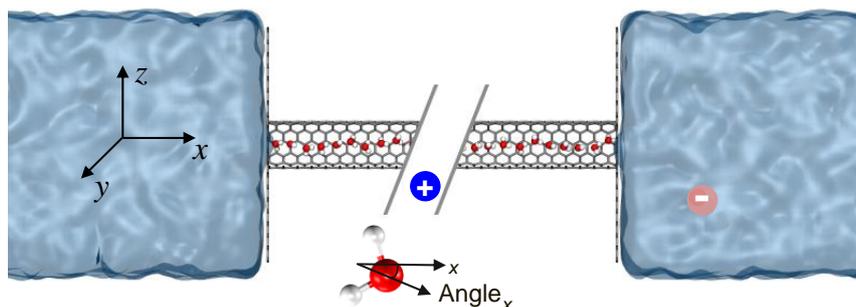
Thus, eqn (3) reduces to Sine-Gordon Equation

$$\phi_{tt} - a\phi_{xx} + b \sin \phi = 0 \quad (5)$$

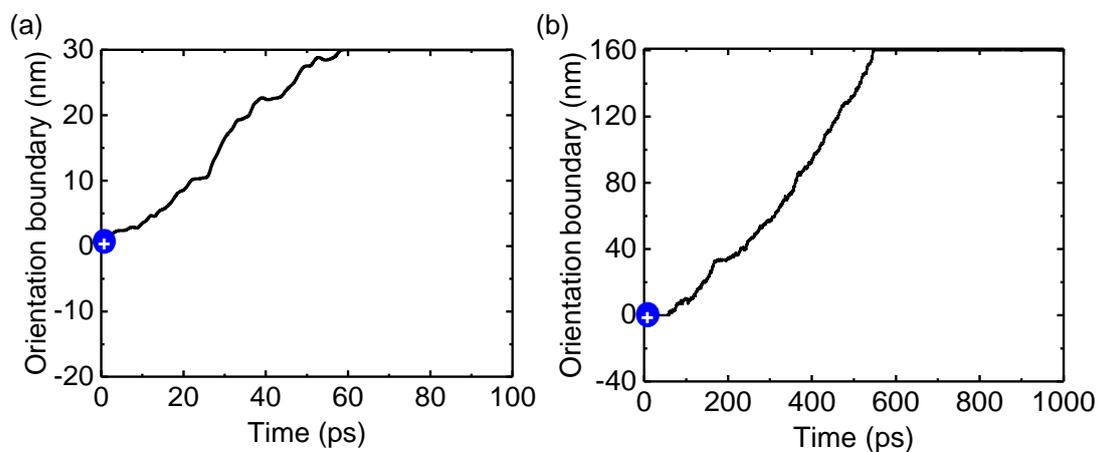
where  $a = \frac{\mu^2}{4\pi\epsilon_0 I r}$ ,  $b = \frac{6.83e_0\mu^2}{2\pi\epsilon_0 I r^3}$ .

## Angle concentration coefficient $\tau$

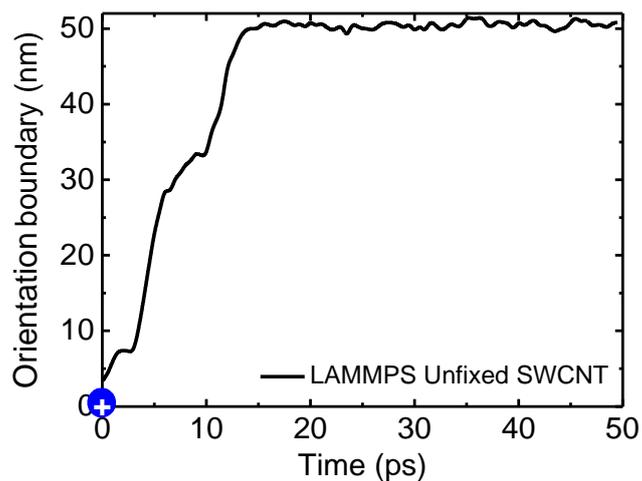
An angle concentration coefficient  $\tau$  was defined to characterize the fluctuation of  $\text{Angle}_x$ , as  $\tau = \frac{N_{<30^\circ}}{N_{total}} / \frac{S_{<30^\circ}}{S_{hemi}}$ , where  $N_{<30^\circ}$  is the number of water molecules with  $\text{Angle}_x < 30^\circ$ ,  $N_{total}$  is the number of all the water molecules in the referred section,  $S_{<30^\circ}$  is the surface area of a unit spherical crown with a vertex angle of  $30^\circ$ ,  $S_{hemi}$  is the surface area of a unit hemisphere. In bulk,  $\tau = 1$ . A larger  $\tau$  corresponds to a smaller fluctuation of  $\text{Angle}_x$  of water molecules.



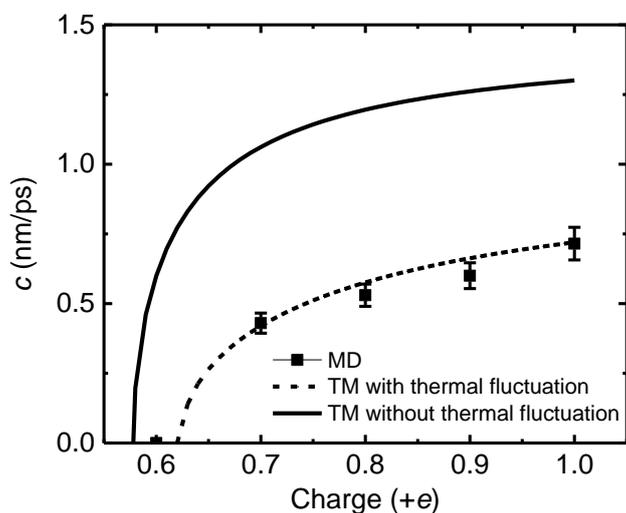
**Fig. S1** A sideview snapshot of the simulation system. Water molecules confined in the SWCNT are drawn in VDW method and the water box is shown partially transparent. The imposed charges are marked by “+” and “-”.



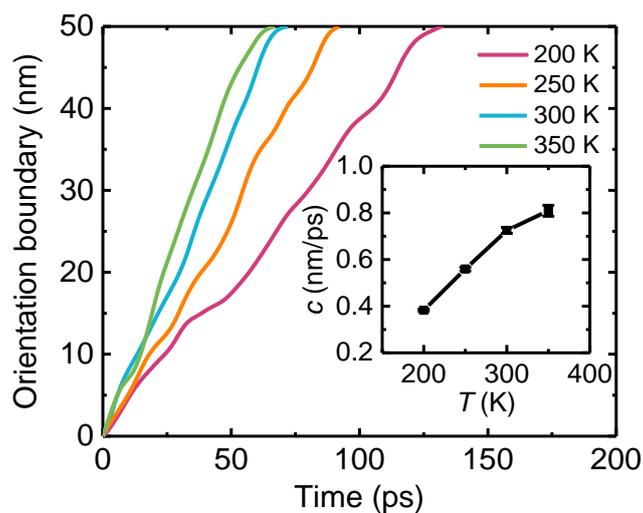
**Fig. S2** Time evolution of the positions of the orientation boundaries in 50 nm (a) and 200 nm (b) long water chains. The location of the charge was set as the origin of the coordinates.



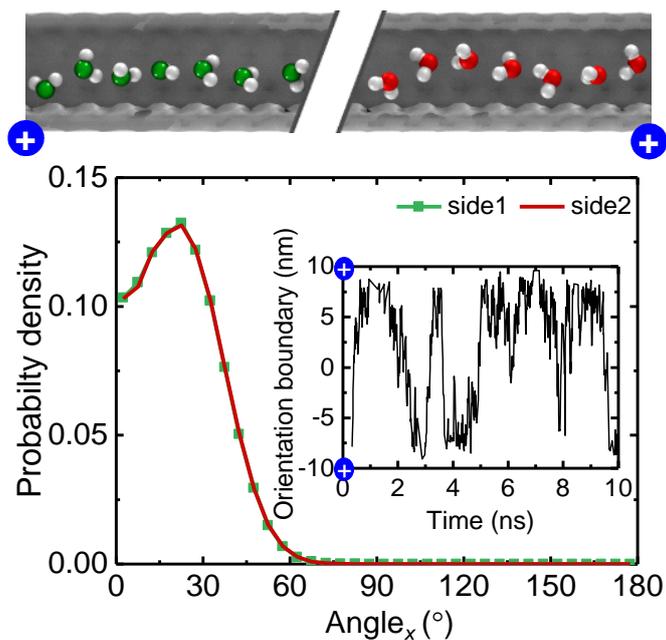
**Fig. S3** Time evolution of the position of the orientation boundary in a water chain. The trajectory was obtained from a simulation carried out using LAMMPS software package with only the terminal atoms of the SWCNT fixed.



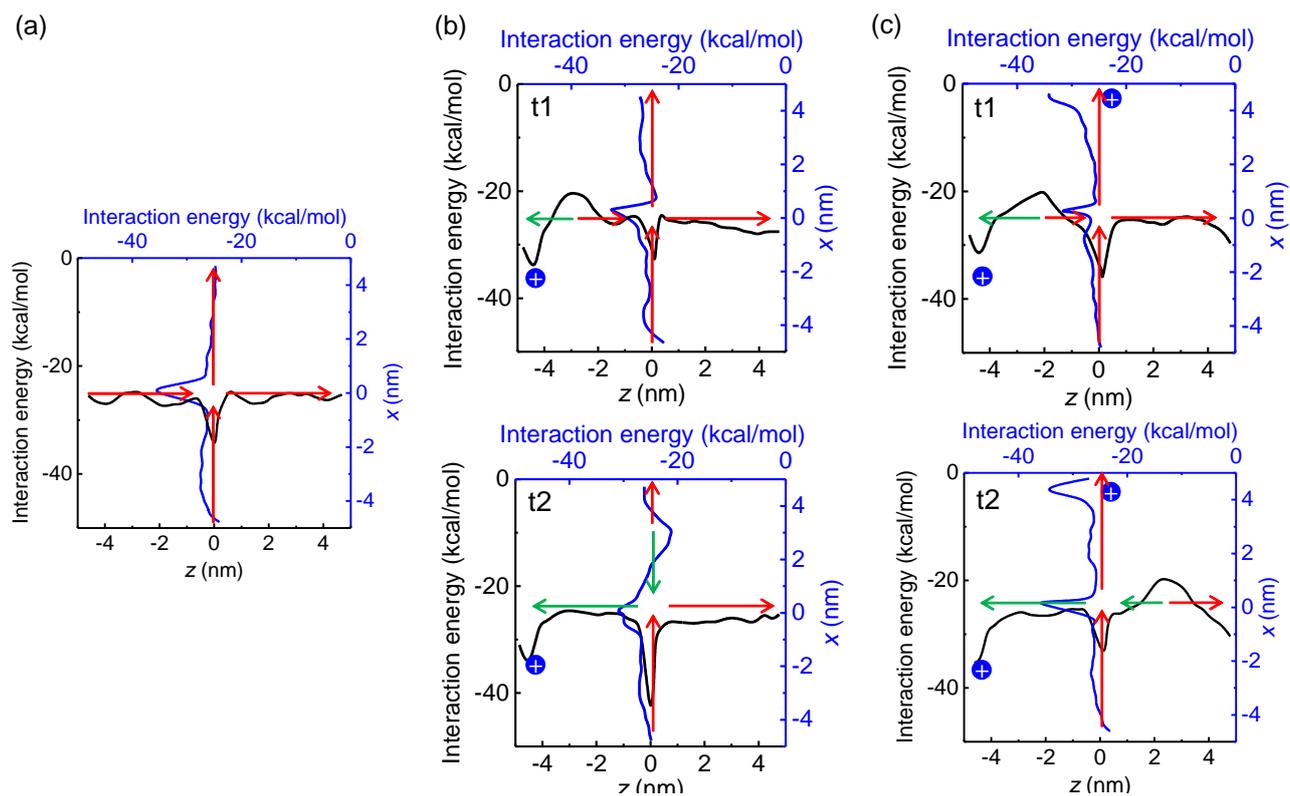
**Fig. S4** Charge dependency of the reorientation propagation velocity  $c$ . The result based on theoretical model (TM) which neglects thermal fluctuations is shown by the solid line, the MD results are shown by cubic symbols and the result based on the corrected theoretical model is shown by the dash line.



**Fig. S5** Time evolution of the orientation boundary along the water chain triggered by an imposed charge  $+e$  at different temperature. Inset shows the propagation velocity  $c$  at different temperature  $T$ .



**Fig. S6** Probability density distribution of Angle<sub>x</sub> of water molecules between two imposed charges  $+e$ . Top panel shows a sketch map. The vibration of the orientation boundary, which divides the water chain into two subsections, namely side1 (green) and side2 (red), is shown in the inset. The midpoint of the two charges is set as the origin of the coordinates.



**Fig. S7** Energy landscape of water chains confined in crossed SWCNTs. (a) Energy landscape of water chains in a system in the absence of imposed charges. The orientations of water dipoles are marked by red arrows. (b) Energy propagation across the crossed junction induced by a positive charge. Energy landscape plots at two typical moments are shown, with the new orientations of water dipoles marked by green arrows. (c) Energy propagation across the crossed junction induced by two positive charges.

**Mov. S1** Reorientation process of water molecules in a single-file water chain.